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STATE-OF-THE-ART. URANIUM MINING, MILLING, AND REFINING INDUSTRY

ROBERT S. KERR ENVIRONMENTAL RESEARCH LABORATORY

PREPARED FOR
OFFICE OF WATER RESOURCES RESEARCH

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STATE-OF-THE-ART: URANIUM
MINING, MILLING, AND REFINING INDUSTRY

by

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The report presents an overview of the uranium mining, milling, and refining industry of the United States. Topics discussed include ore reserves, geographical locations, production statistics, future requirements, processes for extraction and beneficiating, waste characteristics, including radioactive and other potential pollutants, current treatment and disposal methods, effects of wastes on the environment, standards for radiological protection, testing and monitoring programs, technological advances within the uranium industry, anticipated future problems and recommended areas for further study.

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SECTION I

CONCLUSIONS

1. Approximately 95 percent of the known uranium deposits are located within the Western United States; however, the eastern two-thirds has many sedimentary deposits similar to those in the west and exploration activities may uncover new bodies of ore. At present, 40 percent of the United States has potential for uranium ore production.

2. Essentially all of the anticipated uranium requirements in the future will be for a rapidly expanding nuclear power industry utilizing uranium as a fuel source. Uranium requirements are expected to double by 1977, triple by 1981, and continue upward through the year 2000.

3. Exploratory drilling activities have been directed toward finding ore at greater depths with the average depth of drilling increasing from 148 feet in 1958 to 409 feet in 1970. At present, the average grade of uranium ore is 0.22 percent; however, as the richer deposits are depleted in the early 1980's, it will become necessary to mine lower grade ores. Mining and milling of lower grade ores will involve the handling and disposal of significantly larger quantities of waste tailings.

4. Solid waste tailings are accumulating at a rate of approximately 1,960 pounds per ton of ore processed or nine million tons per year from the uranium mills at full operating capacity. By 1972, the total accumulation of tailings in the United States was estimated at 110 million tons containing 60,000 grams of radium-226. Studies have been conducted on various methods of tailings pile stabilization measures against wind and water erosion, but no definitive remedial measures have been set forth as a requirement. Laboratory studies have found that radium-226 may be leached from waste solids up to maximum amounts of 50 percent by contact with water only.

5. Yearly wastes from active uranium mining, milling, and refining plants contain a total of 41,000 curies of radioactivity (6,822 microcuries/ton ore). Radioisotopes with established maximum permissible concentrations contribute 20,500 curies of radioactivity to the waste. The most hazardous of the radioisotopes is radium-226, with a half-life of 1,620 years. Radium is not recovered for commercial uses, as the present world supply is considered adequate. Current production of uranium contributes 3,400 grams of radium yearly to the environment (568 micrograms/ton ore)--more than the estimated total world supply of 3,000 grams. More than 98 percent of the radium is discharged with the solid waste. Other possible contaminants include uranium and other radioisotopes, vanadium, selenium, molybdenum, arsenic, nitrates, sulfates, chlorides, organic extractants, and suspended fines.

6. The few uranium mills currently releasing process waste effluents to rivers and streams treat the waste prior to discharge; however, mine water containing pollutants is being discharged in several mining locations with no chemical treatment. Standard treatment procedure for the removal of radium-226 entails the addition of barium chloride to precipitate radium as a barium-radium sulfate complex. Removal efficiencies of 99 percent may be obtained by the method; however, the process must be carefully monitored or treatment efficiencies decline.

7. For the mills not discharging liquid wastes to the environment, chemical treatment is not practiced; hence, the inorganic and radioactive wastes remain in the dissolved state and are subject to seepage from the tailings ponds. The extent of ground water contamination from this source or the ultimate fate of these pollutants is not known.

8. Comprehensive monitoring programs and background surveys have not been conducted to adequately assess the degree of environmental pollution resulting from uranium mining, milling, and refining operations. With the exception of the Colorado River Basin, data collected from other uranium-producing areas have been insufficient, sporadic, and, in some instances, unreliable.

SECTION II

RECOMMENDATIONS

1. Initiate a comprehensive research program to ascertain the pollutional effects on the environment from all aspects of the uranium mining, milling, and refining industry and to develop adequate waste treatment and control technology for the expanding uranium industry.

2. Research should be conducted to develop new or improved methods to remove radionuclides, vanadium, selenium, molybdenum, arsenic, nitrates, sulfates, chlorides, organic extractants, suspended fines, and other inorganic contaminants from uranium mill process wastes and mine waters regardless of disposal method. Flocculant aids or mechanical means should be employed for better suspended solids removal followed by maximum reuse of the treated waters.

3. A treatment process to remove leachable radium-226 from uranium waste solids prior to disposal should be developed. Countercurrent washing of the tailings solids followed by precipitation or ion exchange concentration might prove feasible. The leaching characteristics of sands and slimes from different types of milling circuits should be studied.

4. Since radium-226 concentrations in uranium concentrates vary according to the milling process utilized, surveys should be conducted to determine the amount of radium-226 passing through the refineries. If levels of the radionuclide are found to be high, treatment of the wastes for removal would be indicated.

5. Physical upgrading processes for uranium ores should be developed and employed to reduce the bulk of solid waste tailings that must be contained and stabilized in restricted areas.

6. The most effective method(s) for the location and stabilization of tailings piles should be determined and uniformly required for all milling

operations. The piles should receive perpetual care with follow-up studies over a period of years to determine efficiency and longevity of the stabilization procedures.

7. Studies should be conducted to ascertain the degree and extent of radioactive and inorganic pollution of surrounding soils and ground waters caused by seepage from uranium tailings ponds and piles. Potential long-range environmental effects could be predicted from this information. Development and utilization of impermeable sealants for tailings ponds would greatly reduce pollution from seepage.

8. A continuous monitoring program should be established for all uranium mining, milling, and refining areas to assure that maximum permissible concentrations of radionuclides and other inorganic constituents are not exceeded. Background surveys to obtain baseline data should be conducted in new areas prior to the initiation of operations.

9. Standards should be established regulating concentrations of pollutants in mine water discharges. A study should be made to determine whether allowable uranium concentrations in water and radium-226 in suspended solids are excessive, and to establish a maximum permissible concentration for molybdenum.

10. Every effort should be made to reduce the concentrations of pollutants released to the environment to the lowest possible level, regardless of the preselected maximum permissible concentrations allowable.

SECTION III

INTRODUCTION

The production of uranium has become a major industry in the United States since the end of World War II. Fission of the atom was successfully demonstrated in 1942, and led the way to the tremendous potential of nuclear power. With the advent of the atomic age, the demand for uranium to be used in the production of nuclear weapons caused extensive exploration for uranium-bearing ore deposits. Major ore deposits are located in the Colorado Plateau encompassing parts of western Colorado, eastern Utah, northeastern Arizona, and northwestern New Mexico. Other extensive deposits are located in Wyoming and the Grants-Ambrosia Lake area of New Mexico. Smaller deposits are located in Texas, North Dakota, South Dakota, and Washington.

By the late 1950's, uranium mills constructed in these areas were processing in excess of 20,000 tons per day of ore with an average uranium oxide (U_3O_8) content of 0.28 percent. The extraction of uranium from the ores leaves large amounts of solid and liquid waste for disposal. Initially, little attention was given to the possibility of significant environmental contamination by the uranium mill wastes. As a result, large quantities of both solid and liquid waste were discarded into streams causing widespread contamination of the affected areas. During the 1950's extensive studies were conducted to determine the fate of radioactive waste materials in the environment and their effects on human health.

Standards for maximum permissible concentrations of radioisotopes in water for human consumption were established; as more was learned about the hazards of radiation, the standards were revised to lower levels.

Methods of waste handling and control were developed in an attempt to lower the pollution to a level that would meet the established standards. The uranium milling industry has implemented pollutional abatement procedures in the plant process to solve some of the problems. With adequate surveillance of the environment, problems can be recognized quickly as they occur, and abatement procedures established to prevent pollution.

A downward trend in uranium production occurred in the early and mid-1960's, as the demand for uranium changed from a military to a smaller commercial market. While nuclear power was feasible, it was not yet economically competitive with other types of fuel. As a result, many mills closed and others reduced their production rate. Exploration activities were curtailed to a greater extent than the milling operations.

Nuclear energy was used solely for military purposes until 1954 when President Eisenhower initiated the "Atoms for Peace" program.[1] Since that time a tremendous amount of effort and money has been spent to utilize nuclear energy for peaceful uses, including electrical power generation, medical diagnosis and therapy, food and materials irradiation, and nonmilitary nuclear explosives.

By 1966 uranium-fueled reactors for production of electric power were developed to the extent that they were economically competitive with other sources of power, thus creating a new market for uranium. Nuclear power generation represented less than one percent of the total electricity generated in 1968, but is expected to provide 40 to 60 percent of the Nation's electricity by the year 2000. In anticipation, a marked increase in exploration for uranium ore deposits to supply future needs has been undertaken.

Advances in technology can be expected in exploration, mining, and processing that will reduce the cost and increase the supply of uranium. Newer methods may pose problems requiring advanced technology in pollution abatement.

SECTION IV

URANIUM ORE SUPPLY

DETECTION OF URANIUM ORE DEPOSITS

In the early days of prospecting for uranium ore, Geiger counters were used to detect deposits within 18 inches of the surface.[2] At the present time, test holes for deeper deposits are drilled and the core measured for radioactivity by means of a scintillation counter or analyzed specifically for uranium by fluorometric or colorimetric methods.

Drilling is the most economical and widely used method of exploration. Most exploratory drilling has been done in districts known to contain uranium deposits. Many new discoveries in these areas are at much greater depths and contain lower grade ore than that previously mined. Exploratory drilling is usually performed using 200 foot centers followed by developmental drilling on 50 foot centers. Small ore bodies have been missed completely when using 100 foot centers. Developmental drilling provides the information needed for mine layout and cost estimates.

Another method of detecting uranium reserves is aerial reconnaissance. The method gives a quick scan of unexplored areas for high radiation zones. The aircraft fly a grid pattern approximately 500 feet above the ground with the grid spacing the same distance apart as the height. Sensitivity is gained by flying closer to the ground, but smaller grid patterns must be made which increase the cost of exploration. Radiation levels obtained may be plotted on a map and the information used for ground instrument surveys or drilling operations to indicate the extensiveness of the deposit.

Monitoring of the ground air for the presence of radon gas is another method that has shown some promise in ore detection.[2] A measurement of the alpha activity is made in auger holes 1.5 to 3 feet deep. This method has been shown to detect radon over uranium-bearing ore covered by considerable thickness of overburden.

ORE RESERVES

The chief sources of ore in the United States are found in sedimentary strata consisting of sandstone, mudstone, and limestone. The uranium minerals occur as pore fillings, replacements of woody tissue or other carbonaceous matter, and as a cement between quartz grains or along fractures.[1]

Uranium deposits occur in either of two modes, stratiform or vein type.[3] Approximately 95 percent of the Nation's reserves is found in the stratiform type consisting of sandstone and conglomerate, limestone, and lignite formations. The deposits lie parallel to the bedding plane and have large lengths and widths as compared to their thickness. Vein-type deposits occurring at steep angles along fractures and structural faults account for the other reserves.

The principal known deposits of uranium ores in the United States are located in three major districts:[4]

1. Wyoming Basins--Shirley Basin, Powder River, and Gas Hills;
2. Colorado Plateau--Uravan Belt, Big Indian, Laguna, and Ambrosia Lake; and
3. Texas Gulf Coast.

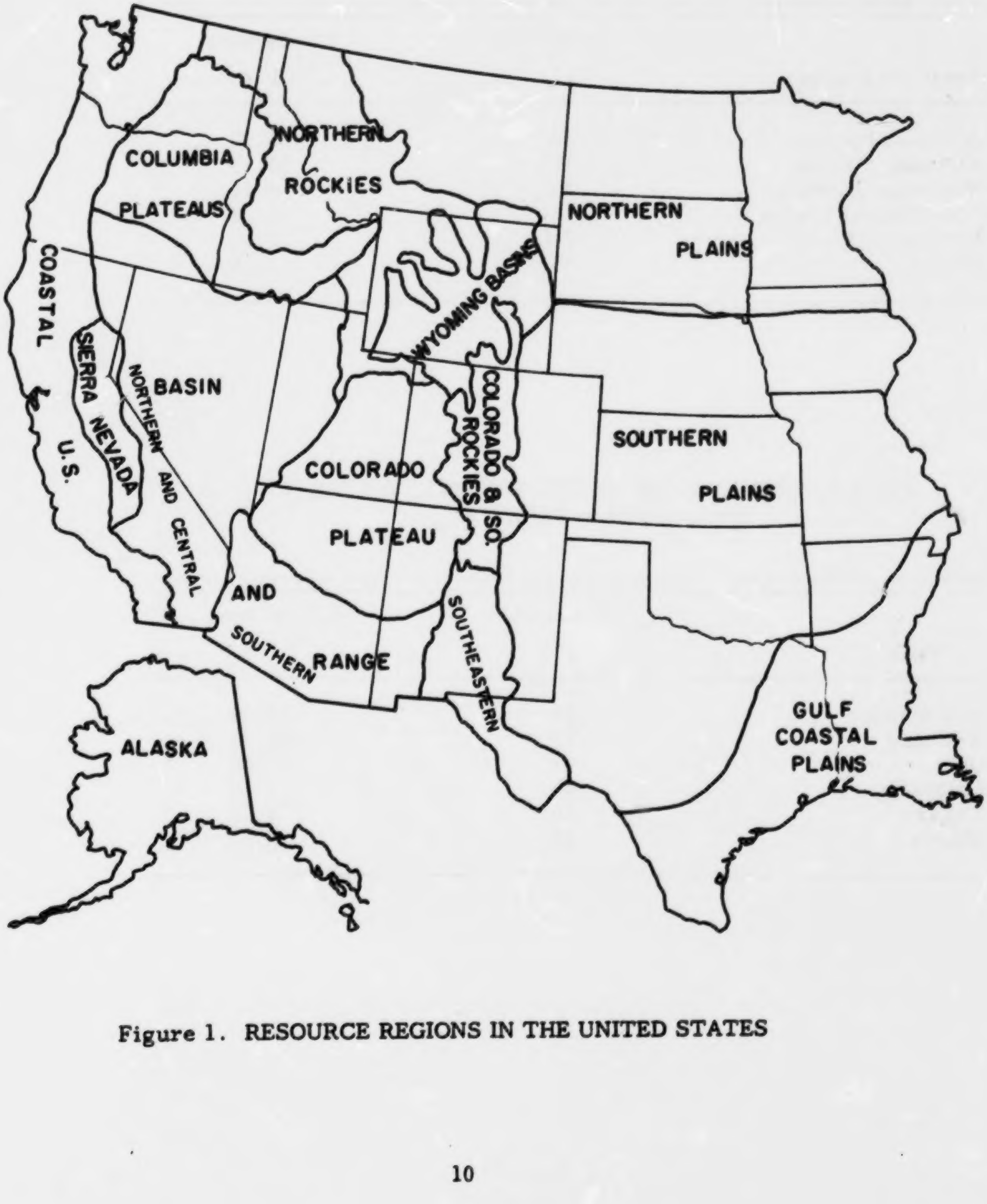
Table 1 shows the percentage distribution of ore reserves by resource region as of January 1971. The reserves are restricted to ore that may be economically produced for $8/lb of U_3O_8. The distribution of ore reserves by State is shown in Table 2, while uranium ore production in the four leading States from 1960 through 1970 is shown in Table 3. Ore reserves now known lie in over a thousand individual deposits in the Western States; however, half of the reserves are in fifteen deposits containing over a million tons of ore.[5] The ore grade ranges from less than 0.1 percent to well over 0.5 percent uranium oxide. Figure 1 shows the geographical location of the ore reserves by resource region. Geographical locations of the major uranium mining districts in the Western States are shown in Figure 2.

Table 1. DISTRIBUTION OF ORE RESERVES BY RESOURCE REGION[7]
(recoverable at \$8/lb U_3O_8)

Resource region	Grade (%) U_3O_8	% Total tons U_3O_8
Colorado Plateau	.24	51.08
Wyoming Basins	.19	38.63
Northern Rockies	.13	4.26
Gulf Coastal Plains	.16	4.22
Northern Plains	.24	0.63
Others	.35	1.18

Table 2. DISTRIBUTION OF ORE RESERVES BY STATE[7]
(recoverable at \$8/lb U_3O_8)

State	Grade (%) U_3O_8	% Total tons U_3O_8
New Mexico	.24	44.04
Wyoming	.19	38.71
Utah	.31	3.68
Colorado	.27	4.14
Texas	.16	4.22
Others	.14	5.21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COLUMBIA PLATEAUS
NORTHERN PLAINS
WYOMING BASINS
COASTAL U.S.
SIERRA NEVADA
NORTHERN AND CENTRAL BASIN AND RANGE
SOUTHERN RANGE
COLORADO PLATEAU
COLORADO & SO. ROCKIES
SOUTHERN PLAINS
SOUTHEASTERN
GULF COASTAL PLAINS
ALASKA

Figure 1. RESOURCE REGIONS IN THE UNITED STATES

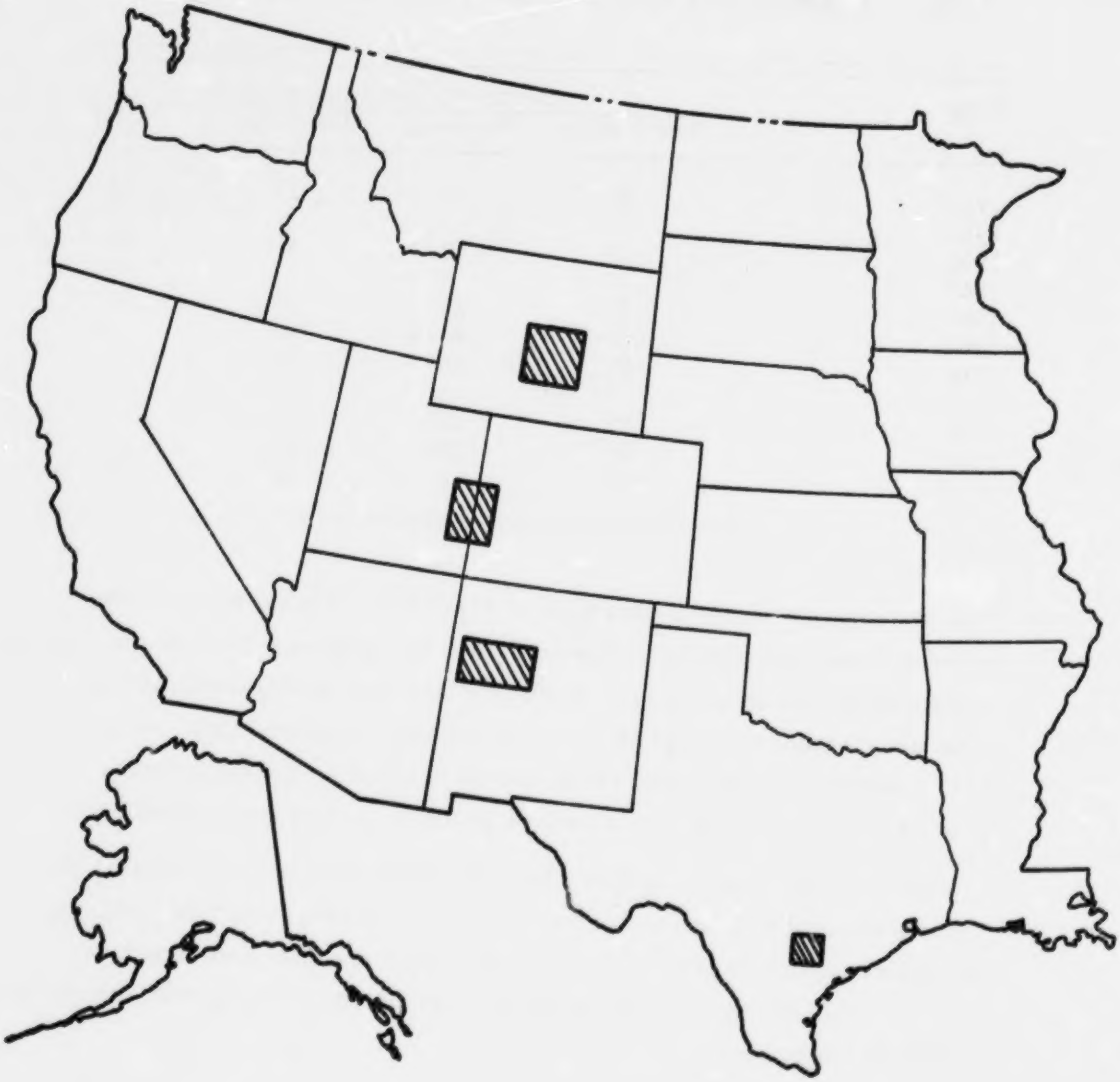

Figure 2. GEOGRAPHICAL LOCATION OF MAJOR URANIUM MINING DISTRICTS

Table 3. URANIUM ORE PRODUCTION IN FOUR LEADING STATES[7, 8]

Year	Percent of total production			
	New Mexico	Wyoming	Colorado	Utah
1960	48	17	14	13
1961	-	-	-	-
1962	50	18	16	11
1963	-	-	-	-
1964	37	26	15	13
1965	46	24	13	9
1966	48	25	15	5
1967	-	-	-	-
1968	51	25	11	7
1969	50	28	12	-
1970	46	26	12	7

The ore deposits in the Texas Coastal Plain were discovered in 1954, when a plane carrying scintillation detection equipment recorded strong anomalies in Karnes County.[6] Probably less than five percent of the total prospectable acreage in the area had been explored as of January 1967. Some deposits of ore are expected to occur in the High Plains area of Texas, but will likely be less productive than the Coastal Plains.

Lignite ores are located in North Dakota, South Dakota, and Texas. The Uravan mineral belt in the Colorado Plateau contains vanadium-bearing minerals, making it profitable to recover both uranium and vanadium from the ore. Much of the uranium ore contains molybdenum in quantities sufficient for economic recovery.

Canadian companies are conducting exploration activities in the United States.[9] Rio Algom Mines Limited is developing a mine in the Lisbon Valley district, about 30 miles southeast of Moab, Utah, and is participating in an extensive exploration program in Wyoming with Mitshubishi Metal Mining Company. Several major uranium-producing companies from the United States are becoming involved in Canadian uranium exploration activities in anticipation of future uranium requirements.

Many new companies are entering the field of uranium exploration; the number of companies exploring for uranium more than doubled from 1965 through 1967. The discovery rate from exploratory drilling in the previous uranium-producing areas has been about six pounds of uranium oxide per foot drilled. Recently, exploration has spread into previously unexplored areas and, combined with the search for deeper deposits, has resulted in a reduction in the discovery rate to about three pounds of uranium oxide per foot drilled.[10]

Uranium has been found and produced in 17 States, but 95 percent has been from Arizona, Utah, New Mexico, and Wyoming.[11] The eastern two-thirds of the United States has many sedimentary deposits similar to those in the uranium-producing areas. As exploration activities spread into the newer areas, new bodies of ore may be discovered to greatly expand reserves. About 40 percent of the United States has potential for producing uranium ore.

Drilling activities have increased rapidly since 1967. Table 4 shows the surface drilling from 1967 through 1970 with the projected drilling plans for 1971 through 1973. The previous peak in drilling was in 1957 when 9.2 million feet were drilled. In comparison, drilling from 1951 through 1960 was only 52 million feet.

Table 5 shows both exploratory and surface drilling for the period 1966 through 1970. The exploratory drilling has increased at a much faster rate than developmental drilling, indicating a rush to find new reserves for future demands. The previous high in exploratory drilling was in 1957 when 7.3 million feet were drilled.

Table 6 shows the distribution of the 1970 surface drilling by States. Of interest is the high drilling activity in Texas for new reserves. While producing only five percent of the uranium, it ranks second in drilling activity. This may be an indication of a forthcoming rise in production rate.

Table 4. SURFACE DRILLING[7]

Year	Million feet
1967	10.7
1968	23.8
1969	29.9
1970	24.0
1971-1973	54.2 (projected drilling plans)

Table 5. DRILLING ACTIVITY[7]

Year	Exploratory	Developmental
1966	1,800,000 ft.	2,400,000 ft.
1967	5,435,000 ft.	5,329,000 ft.
1968	16,227,000 ft.	7,527,000 ft.
1969	20,470,000 ft.	9,385,000 ft.
1970	17,981,000 ft.	5,547,000 ft.
1971	12,200,000 ft. (estimate)	7,000,000 ft. (estimate)
1972	11,600,000 ft. (estimate)	6,700,000 ft. (estimate)
1973	9,900,000 ft. (estimate)	6,800,000 ft. (estimate)

Table 6. DISTRIBUTION OF THE 1970 SURFACE DRILLING BY STATES[7]

State	Drilling	% of Total
Wyoming	9,812,000	41.7
Texas	6,075,000	25.8
New Mexico	5,180,000	22.0
Colorado	1,007,800	4.3
Utah	641,300	2.7
South Dakota	436,900	1.9
Others	374,900	1.6
Total	23,528,000	100.0

A trend toward deeper drilling has taken place since 1965. Shallow deposits were more readily located by airborne or ground surveys and more economical to mine. As these deposits become depleted, exploration at deeper depths will be required. Table 7 shows the drilling statistics for the Western United States. From 1960 through 1965 the average depth per hole drilled was 160 feet. The average depth of drilling has steadily increased through the years from 148 feet in 1958 to 409 feet in 1970. From 1965 to 1970 the average depth more than doubled. Table 8 shows the distribution of ore by depth for the year 1970. Approximately 65 percent of the ore reserves lie at depths of less than 500 feet allowing recovery by open pit mining. The ore is more concentrated at two zones: 150 to 200 feet and 350 to 400 feet where 14.3 and 16.2 percent of the ore is found.

Table 9 shows the distribution and grade of ore reserves by mining method as of January 1971. Open pit mines are located in Wyoming, New Mexico, and Texas. Underground mines are located in all the uranium-producing States except Texas.

As early as 1957, large ore reserves had been developed, but as the requirements for uranium decreased, production experienced a downward trend from 1960 through 1966. Exploration activities were halted and many mills were closed. As the nuclear power industry began to grow, increased exploration activity in 1967 resulted in more new reserves discovered than those mined in that year. The reserve estimation from 1955 through 1970 is shown in Table 10.

Should the market price rise substantially, reserves can be increased by mining lower grade ore containing as little as 0.03 percent uranium oxide. Phosphates in Florida have a uranium content of 0.005 to 0.03 percent uranium oxide. Marine shales range from 0.001 to 0.02 percent uranium oxide and are found in Kentucky, Tennessee, and Alabama. New technology in mining and milling processes may make mining of low grade

Table 7. DRILLING STATISTICS FOR WESTERN UNITED STATES[7]

Year	Average depth (ft.)	
	Exploratory	Developmental
1958	148	152
1959	146	168
1960	191	173
1961	160	165
1962	230	189
1963	104	146
1964	162	126
1965	187	129
1966	313	182
1967	425	314
1968	422	385
1969	428	335
1970	409	373

Table 8. DISTRIBUTION OF 1970 URANIUM OXIDE PRODUCTION IN ORE BY DEPTH OF ORE[7]

Feet	Percent
0- 50	2.47
50- 100	4.90
100- 150	8.13
150- 200	14.30
200- 250	4.10
250- 300	3.65
300- 350	0.51
350- 400	16.18
400- 450	2.57
450- 500	1.46
500- 550	8.22
550- 600	0.90
600- 650	6.94
650- 700	6.90
700- 750	6.15
750- 800	5.62
800-1,500	4.67

Table 9. DISTRIBUTION OF ORE RESERVES BY MINING METHOD[7]

Mining method	Grade (%) U_3O_8	% Total tons U_3O_8
Open Pit	.19	53.72
Underground Mines	.24	46.23
Others[a]	.13	0.05

[a] Heap and in situ leaching, extraction from mine water.

Table 10. URANIUM ORE RESERVES AND PRODUCTION
1955 THROUGH 1970[7]

Year end	Shipment to mills[a] tons of U_3O_8	Reserve estimation tons of U_3O_8
1955	4,425	67,595
1956	8,434	164,055
1957	9,837	210,109
1958	14,003	225,644
1959	17,377	240,996
1960	18,842	231,785
1961	18,513	178,385
1962	17,085	167,738
1963	14,721	160,231
1964	13,888	150,927
1965	10,578	144,702
1966	10,051	140,835
1967	10,866	147,741
1968	12,850	160,819
1969	12,595	204,080
1970	13,073	246,100

[a] Includes miscellaneous U_3O_8 receipts from mine waters, heap leach, in situ, and refining residues.

minerals economical without greatly raising the market price. The leaching of uranium from low grade copper dumps and recovery from phosphate fertilizer is projected to supply 100,000 tons of uranium oxide through the year 2000.[10]

Another source of uranium may be the surplus stockpiled uranium concentrate held by the Atomic Energy Commission.[5] The AEC stopped procurement at the end of 1970 with 40,000 to 50,000 tons of stockpiled uranium oxide. Release of the surplus for commercial use may begin in 1975, but be controlled so as to protect uranium producers. Should a shortage develop due to nuclear power demands, the surplus could serve to alleviate the situation.

EFFECT OF THE NUCLEAR POWER INDUSTRY ON URANIUM REQUIREMENTS

The introduction of uranium-fueled nuclear power generators has created a new market for uranium. Nuclear energy for the production of electricity has become economically competitive with other types of energy.

It is believed that practically all of the demand for uranium for non-military purposes to the end of the century will be to fuel nuclear power reactors. Uranium's greatest advantage as a fuel for the production of electricity is that an enormous amount of energy is stored in a comparatively small space. One pound of uranium has the same energy potential as three million pounds of coal. After years of development by government and industry, the manufacturing facilities in the United States are capable of producing an estimated 20 large nuclear power reactors each year.[15]

During 1970, five new nuclear power reactors began operations with 14 more under contract, making a year-end total of 108 central station nuclear power reactors under contract, under construction, or operable in the United States. It is predicted that penetration of the electric utility market by nuclear power plants will increase from less than one percent of the total generation in 1968, to 25 to 30 percent in 1980, and range from 40 to 60 percent in the year 2000.[1, 12]

It is expected that breeder reactors will be developed by the mid-1980's. Only one percent of the uranium-235 portion of the uranium oxide concentrate is consumed in the nuclear reactors presently in use. In contrast, the breeder reactor will actually produce more fuel than it consumes.[13] The breeder reactor will produce fissionable plutonium-239 and uranium-233 from uranium-238 and thorium-232, respectively. The ultimate effect of utilizing the breeder reactor will be to reduce the amount of uranium required, thereby increasing the currently predicted uranium depletion times from decades to centuries. Another advantage is that the breeder reaction is relatively insensitive to cost of uranium oxide, thus allowing mining and milling lower grade ores at a higher market price to provide additional ore reserves.

The Liquid Metal Fast Breeder Reactor has been chosen by the Atomic Energy Commission as the prime contender for reactors of the future and a development program is under way to perfect its design.[14] This type of reactor should compare economically with reactors now in operation, and use the nuclear resources more efficiently than present reactors. Additionally, it will be able to operate on reprocessed spent fuel from present reactors, thus eliminating uranium isotopes and plutonium-239 from the current and perplexing waste disposal problem.

Should breeder reactors be introduced in the early 1980's, the effect on the uranium requirements will probably not be evident until the end of the century. Should plutonium-239 be reserved for breeder fuels, the demand for uranium to fuel light water reactors would increase. Also a large amount of uranium will be required for the initial fueling of these new reactors.

Plutonium recycle in thermal reactors is planned to start in 1974, thus reducing annual requirements. Many uncertainties in requirements for the late 1980's and the 1990's exist, such as the timing of commercial fast breeder reactors. A substantially greater amount of uranium will be required, but a close estimate is difficult at this time. Table 11 shows the projected domestic uranium requirements through the year 2000. The requirements include the initial fuel for reactors under construction and makeup fuel for operating reactors. Requirements for operating reactors ranged from 15 percent of the total annual requirements in 1966, to 60 percent in 1980.

Current figures for the number of nuclear power plants to the end of the century indicate a steeply rising annual demand for uranium far beyond the capacity of existing mills. Existing mills produced 13,073 tons of uranium oxide in 1970, as seen from Table 10. The projected annual requirements will reach this level in 1973, as seen in Table 11, will double by 1977, and triple by 1981. New mines and processing mills will have to be placed in operation if the projected requirements are to be met. An eight-year forward reserve is needed to assure an adequate supply of ore for the requirements; hence, each year the addition to the reserves must equal the annual requirements eight years later.

MINING AND MILLING OPERATIONS

The number of operating mines has steadily decreased from 1,000 in 1960 to 310 in 1969, as shown in Table 12. Many of the earlier mines were one-man operations producing small quantities of ore. The ore was hauled from the many locations to a processing mill some distance away. The ore production rate in 1969 was 75 percent of that in 1960, despite the fact that the operating mines had decreased to 30 percent of the 1960 figure. This was due to the larger companies taking over the majority of the smaller mining operations.

There were 25 mills in operation in 1960 that produced 17,646 tons of uranium concentrate. The number of mills decreased to 15 in 1969, due to the lowered demand. In anticipation of the increased demand for uranium as a fuel for nuclear power, two new mills were placed in

Table 11. URANIUM REQUIREMENTS[1, 4, 5, 7, 9, 11, 16]

Year	Annual tons of U_3O_8	Cumulative tons of U_3O_8
1968	4,000	4,000
1969	5,000	9,000
1970	7,000	16,000
1971	8,700	24,700
1972	10,500	35,200
1973	13,000	48,200
1974	16,000	64,200
1975	18,500	82,700
1976	21,500	104,200
1977	25,000	129,200
1978	28,000	157,200
1979	32,000	189,200
1980	36,000	225,200
1981	41,000	266,200
1982	46,000	312,200
1983	51,000	363,200
1984	56,000	419,200
1985	60,000	479,200
1986	62,500	541,700
1987	65,000	606,700
1988	67,000	673,700
1989	68,500	742,200
1990	69,500	811,700
1991	70,500	882,200
1992	71,000	953,200
1993	72,000	1,025,200
1994	72,500	1,097,700
1995	73,200	1,170,900
1996	74,000	1,244,900
1997	75,000	1,319,900
1998	75,500	1,395,400
1999	76,500	1,471,900
2000	77,000	1,548,900

Table 12. URANIUM MINES AND ORE-PROCESSING PLANTS[8]

Year	Operating mines	Tons of ore produced	Operating mills	Tons of U_3O_8 concentrate to AEC	Tons of U_3O_8 concentrate to private industry
1960	over 1,000	8,000,000	25	17,646	-
1961	over 1,000	8,000,000	26	17,348	-
1962	over 1,000	7,000,000	24	17,010	-
1963	730	5,900,000	24	14,218	-
1964	600	5,700,000	21	11,847	-
1965	650	4,400,000	19	10,442	-
1966	-	4,352,651	17	9,487	-
1967	500	5,300,000	16	8,425	700
1968	320	6,500,000	16	7,338	5,000
1969	310	5,900,000	15	6,184	6,400
1970	-	-	17	2,500	11,200
1971	-	-	17	-	-

operation in 1970. One of these was Susquehanna-Western, Incorporated, located near Three Rivers, Texas; the other was Utah Construction and Mining Company at Shirley Basin, Wyoming. Two more acid leach-solvent extraction plants were placed in operation in 1972. Continental Oil Company opened a mill near Falls City, Texas, with a capacity of 1,750 tons of ore per day, and Humble Oil and Refining Company began operations at a mill near Douglas, Wyoming, with a capacity of 2,000 tons of ore per day. An alkaline leach plant with a 500 tons of ore per day capacity was also placed into operation in 1972 by Rio Algom Corporation near La Sal, Utah. Table 13 lists operating mills, location, and capacity at the end of 1972.

At one time 34 mills were located in the Western United States with 17 located within the Colorado River Basin. The locations of the Colorado River Basin mills are shown in Table 14. Only three of the mills are still in operation. Although operations have ceased at 14 of the mills, waste tailings piles containing potential radioactive pollutants remain at the sites.

Table 13. URANIUM ORE-PROCESSING PLANTS 1972

State and company	Plant location	Nominal capacity (tons ore per day)
Colorado:		
Cotter Corp.[a]	Canon City	450
Union Carbide Corp.	Rifle	2,000
Union Carbide Corp.	Uravan	
New Mexico:		
The Anaconda Co.	Bluewater	3,000
Kerr-McGee Corp.	Grants	7,000
United Nuclear-Homestake Partners	Grants	3,500
South Dakota:		
Mines Development, Inc.[a]	Edgemont	650
Texas:		
Susquehanna-Western, Inc.[a]	Falls City	1,000
Susquehanna-Western, Inc.	Three Rivers	1,000
Continental Oil Company	Falls City	1,750
Utah:		
Atlas Corp.	Moab	1,500
Rio Algom Corporation	La Sal	500
Washington:		
Dawn Mining Co.[a, b]	Ford	500
Wyoming:		
Federal American Partners	Gas Hills	950
Petrotomics Co.	Shirley Basin	1,500
Union Carbide Corp.	Gas Hills	1,000
Utah Construction & Mining Co.	Gas Hills	1,200
Utah Construction & Mining Co.	Shirley Basin	1,200
Western Nuclear, Inc.	Jeffrey City	1,200
Humble Oil and Refining Co.	Douglas	2,000
	TOTAL	31,900

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[a] Private sales only. Mines Development, Inc. shipped a small quantity to AEC.

[b] Reactivated early in 1970.

Table 14. COLORADO RIVER BASIN URANIUM MILLS

Mill location	Began operation
Rifle, Colorado (old mill)	1947
Rifle, Colorado (new mill)	1958
Uravan, Colorado	1949
Naturita, Colorado	1947
Slick Rock, Colorado	1958
Maybell, Colorado	1957
Gunnison, Colorado	1958
Durango, Colorado	1949
Grand Junction, Colorado	1951
Moab, Utah	1956
Mexican Hat, Utah	1957
Monticello, Utah	1949
Hite, Utah	1949
Green River, Utah	1958
Monument Valley, Arizona	1955
Shiprock, New Mexico	1954
Tuba City, Arizona	1956

SECTION V

MINING PROCESSES

OPEN PIT MINING

Open pit mining accounts for slightly more than half of the reserves mined, and uncovers more ore per mine than does underground mining. In 1969 there were 25 open pit mines and 214 underground mines being worked.[3]

Open pit mining is used where the ore deposits are near the surface and covered with loose, easily removable soil. Some open pit mining may be done at depths of more than 500 feet; but usually, below 300 feet, underground methods are preferred.[1] The ratio of overburden to ore removed in uranium mines is unusually large as compared to other types of mining with ranges from 8: 1 to 35: 1. The expense of removing the larger amounts of overburden is justified by the greater value of the product being recovered.

Conventional earth-moving equipment is used for mining: scraper-loaders, bulldozers, gas and diesel shovels, and rippers. The size of the mining operation determines to some extent the equipment employed. In some small ore bodies backhoes are the most economical means of digging and loading ore.

Ground water intrusion has been a problem in many of the open pit mines. The water is pumped from the mine to keep the floor surface workable. A trench several feet deep may be dug around the periphery of the pit floor and as the ground water drains from the pit floor into the ditch, it is pumped from the mine. As the ore is removed to the level of the ditch depth, the process is repeated. The water is discharged to the surface to seep into the ground or drain into nearby creeks or rivers.

UNDERGROUND MINING

Underground mining is carried out in Utah, Colorado, Wyoming, and New Mexico with Utah and Colorado supporting three-fourths of the producing mines, many of which are small. About 70 percent of the New Mexico production is underground. Underground mining is done on a small scale in the Gas Hills and Crooks Gap District of Wyoming.

The largest ore bodies mined by underground methods measure as much as half a mile in length, several hundred feet in width, from 5 to 100 feet thick, and are located several hundred feet below ground. Many of the smaller deposits are mined using simple adits in canyon walls with removal of ore by wheelbarrows. Most mines require shaft entry, but some ore is mined using inclines and adits. Shafts, located at depths of 800 feet, are generally concrete-lined for lowered maintenance. Stoping methods are generally used with various forms of room and pillar methods. When possible, waste or low grade ore is left as pillars. Tunnels extending from the shaft are supported by steel plate, timber, or concrete, depending on ground conditions and permanency of the tunnel.

The ore bodies are outlined by underground long-hole drilling of 100 to 300 feet from the underground shaft and tunnels. New tunnels are placed from the long-hole drilling data. The mine is continually developed in this manner until the vein is depleted. Ground water from the ore bodies is pumped to the surface for discharge or used as processing water in the mill. The volume of water pumped from mines may range from 200 to 3,000 gallons per minute.

Lack of proper ventilation in uranium mines constitutes a hazard since radon-222, a radioactive gas, is produced as one of the daughter products of uranium. Without adequate ventilation the gas will concentrate in levels considered hazardous to the miners' health.[17, 18] Daughter products of radon-222 are solids that may deposit on the lung surfaces until decay. Concern has been expressed that uranium miners may be subject to an increased risk of lung cancer due to radon-222 and its daughters. Epidemiological studies of lung cancer among miners have been under way by the U.S. Public Health Service for some years.[19] Fans are installed to circulate the air and large-diameter vent holes are drilled to get sufficient fresh air into the mine.

SOLUTION MINING

Solution mining or in-place leaching is a process by which an acid solution is pumped into the underground ore body, allowed to solubilize the desired element, and the pregnant solution recovered by pumping to the surface. Uranium is extracted from the solution by chemical means.

Since 1963 the Utah Construction and Mining Company has been using a solution mining process routinely to extract uranium from an underground ore body in Shirley Basin, Wyoming.[20] Certain conditions must exist for solution mining of uranium:

1. Uranium ore must lie in a generally horizontal bed underlain by a relatively impermeable stratum.
2. The ore must occur below the static water table.
3. The direction and velocity of regional waterflow must be known.
4. Mineralogy of the ore must be determined to choose the proper leaching and extracting process.

The well locations and the inflow-effluent rates must be carefully planned using regional water flow data and experimental drilling. The Shirley Basin mining operation is composed of three inflow wells upgradient from a production well with the center inflow well directly upgradient on the regional ground water flow direction. The remaining two inflow wells are located on radii diverging at an angle of 75 degrees from one another, equally spaced from the center inflow well. The distance from the inflow wells to the production well is approximately 25 feet.

A five percent sulfuric acid solution is pumped into the inflow wells at a slightly slower rate than is being withdrawn from the production well. Sodium chlorate may be added as an oxidant to increase the leaching efficiency. The flow of solution is continued until the concentration of uranium in the leach solution decreases, indicating the leach zone is depleted. Approximately one month is required for depletion of a zone with three to five well patterns in operation simultaneously. Uranium recovery approaches 100 percent except when multiple-horizon areas occur, and upper horizons are not underlain by impervious layers.

The uranium-bearing liquor, containing between 0.10 and 0.30 grams uranium oxide per liter, is passed through an ion exchange column for adsorption on a resin. The loaded column is stripped with a mixture of nitric acid, sodium nitrate, and sulfuric acid. The barren leach solution is discharged as waste to a tailings pond with no outflow. The strip solution is then precipitated with a lime or magnesium slurry at pH 7.5; the precipitate allowed to settle, and separated by decanting. The slurry is shipped to the mill for further purification. The decant, containing neutral nitrate salt, is recirculated to eluate makeup with nitric acid.

Solution mining costs approach or exceed those of open pit mining but are lower than those of underground mining. The operation is much safer for mining personnel, however. Disadvantages are that the proper ground conditions for recovery must be present for utilization of the process. Also, in poor recovery zones, injected solution may be lost in multiple ore horizons and underground fractures, thus becoming a contaminant to ground waters. Solution mining can be used on mined-out stopes and tunnels of underground mines by flooding with leach solution and then pumping the loaded solution out of the mine for extraction of the uranium. Natural leaching of uranium ore by the ground water results in uranium concentrations containing up to ten parts per million that may be extracted by ion exchange processes.

Bacterial leaching, a solution mining technique, involves the oxidation of pyrite (FeS_2) in the presence of water to form ferrous sulfate and sulfuric acid. Certain acid tolerant bacterial species are capable of further oxidizing ferrous sulfate to ferric sulfate, which in turn oxidizes the insoluble tetravalent uranium to the acid soluble hexavalent state. In practice, mines are flooded with water which gradually decreases in pH to between 1.8 and 3.5 as sulfuric acid is formed. The pregnant mine water is pumped to an ion exchange extraction circuit for uranium separation.

Studies in Canada have been conducted to determine whether the bacterial leaching process could be used in the mill with run-of-mine ore.[21] Sulfuric acid requirements for leaching were reduced from 80 pounds per ton to 25 pounds per ton. Underground bacterial leaching has been used in Canada with success, but is unlikely to supersede conventional mining and milling methods due to the long period of time required for efficient leaching.[22] The chief uses have been to scavenge worked-out mines, caved areas, or low grade materials above or below ground that are uneconomical to treat by conventional methods. The process also holds promise for the recovery of uranium from material rejected by flotation, heavy media, electronic sorting, or other upgrading processes. The bacterial leaching of United States ores may never be effective because most presently known reserves contain insufficient pyrite and an abundance of neutralizing calcium carbonate.

Stockpiles of low grade ore removed from mines may be profitably processed by heap leaching.[4, 23] Heap leaching does not require a large capital expenditure for equipment and manpower requirements are minimal. Heap leaching has been practiced by the Utah Construction and Mining Company and Western Nuclear, Incorporated in Wyoming.

A typical heap leaching pile is constructed by grading the ground at the site area to a smooth sloping surface. The area, approximately 300 feet wide by 700 feet long, is covered by a polyethylene sheet of six mil thickness. Four-inch perforated plastic pipe is placed parallel to the width at 18 foot centers. The pipe is covered with one foot of gravel followed by emplacement of low grade ore to a depth of approximately 25 feet. The top of the pile is graded and divided into sections of 300 feet by 60 feet with dikes made from the ore. A sulfuric acid solution is placed in the diked sections of the pile, allowed to percolate through the ore, collected by drainage from the pipes, and removed to storage tanks. The uranium is extracted from the leach solution by conventional solvent extraction or ion exchange methods. Waste acidic solutions are recycled through the pile for maximum leaching efficiency. The final strip solution contains approximately 25 grams of uranium per liter that may be further processed by a mill. Airborne radiation surveys are made during the operation of the system to check for radon-222 in the air. The pile, containing approximately 250,000 tons of ore, is abandoned as leaching operations are completed. The residual material contains less than 0.05 percent uranium. Western Nuclear's facility produces approximately 12,000 pounds of uranium per month at the Day Loma heap leach site.

SECTION VI

MILLING PROCESSES

The uranium recovery process varies from mill to mill depending upon the presence of undesirable constituents in the gangue of the ore or other valuable constituents to be recovered. The milling process may be broken down into separate circuits; each of the operating mills is composed of various combinations of circuits as shown in Table 15. The organic extractants and precipitating agents utilized are listed in the respective columns. Uranium precipitates containing sodium impurities are further purified by acid dissolution followed by reprecipitation with ammonia. As far as possible, the process solutions are recovered and recycled in the milling processes. The remainder is discharged as waste to the disposal area along with leached sands and slimes and dissolved constituents from the ore.

Flow diagrams of various milling processes are shown in Figures 3 through 9. The flowsheets reflect only the sand-slime discharges to the tailings pond. Recycling of process solutions within the plant is not shown because of mill variations in volume, chemical content, and point of discharge to tailings pond. Unrecycled process solutions are used to carry sand and slimes to the tailings area. Additional water is added as necessary to supplement process solutions in slurrying the solids for transport to the tailings pond. Excessive chemical buildup in the recycled mill solutions from the dissolution of minerals in the ore is prevented by discharging a portion of the solution to the tailings pond and replenishing that volume with additional water and chemicals. A number of mills reuse tailings pond water following removal of the suspended solids.

Table 15. URANIUM MILLING PLANTS AND PROCESSING OPERATIONS

Mill and location	Processes used[a]					
	Leaching process	Liquid-solid separation	Ion exchange	Solvent extraction	Eluex process	Precipitation
Colorado:						
Cotter Corp. (dual process)	AL	Drum filters	---	EHPA[b]	---	---
	ALKL	Drum filters	---	---	---	NaOH, H_2SO_4, NH_3
Union Carbide Corp., Rifle	AL	CCD	---	EHPA	---	NaOH
Union Carbide Corp., Uravan	AL	CCD	IX	---	---	NH_3
New Mexico:						
The Anaconda Co.	AL	CCD	RIP	---	---	MgO
Kerr-McGee Corp.	AL	CCD	---	AMINE	---	NH_3
United Nuclear-Homestake Partners	ALKL	Drum filters	---	---	---	NaOH, H_2SO_4, NH_3
South Dakota:						
Mines Development, Inc.	AL	CCD	RIP	EHPA	Yes	H_2O_2
Texas:						
Susquehanna-Western, Inc., Falls City	AL	CCD	---	AMINE	---	NH_3
Susquehanna-Western, Inc., Three Rivers	ALKL	Drum filters	---	---	---	NH_3
Utah:						
Atlas Corp. (dual process)	AL	CCD	---	AMINE[c]	---	---
	ALKL	---	RIP	---	---	NH_3
Washington:						
Dawn Mining Co.	AL	CCD	RIP	---	---	NH_3
Wyoming:						
Federal American Partners	AL	CCD	RIP	AMINE	Yes	NH_3
Petrotomics	AL	CCD	---	AMINE	---	NH_3
Union Carbide Corp.	AL	CCD	RIP	SX	Yes	NH_3
Utah Construction and Mining Co., Gas Hills	AL	CCD	IX	AMINE	Yes	NH_3
Utah Construction and Mining Co., Shirley Basin	AL	CCD	IX	---	---	NH_3 or MgO
Western Nuclear, Inc.	AL	CCD	RIP	AMINE	Yes	NH_3

[a] AL - Acid Leach
ALKL - Alkaline Leach
CCD - Countercurrent Decantation
RIP - Resin in Pulp
IX - Column Ion Exchange
SX - Solvent Extraction

NaOH - sodium hydroxide
H_2SO_4 - sulfuric acid
NH_3 - ammonia gas
MgO - magnesium oxide
H_2O_2 - hydrogen peroxide

[b] The loaded strip solution is combined with the filtered alkaline leach solution from the alkaline mill process for precipitation.

[c] The loaded strip solution is combined with the loaded strip solution from the alkaline-RIP circuit for precipitation.

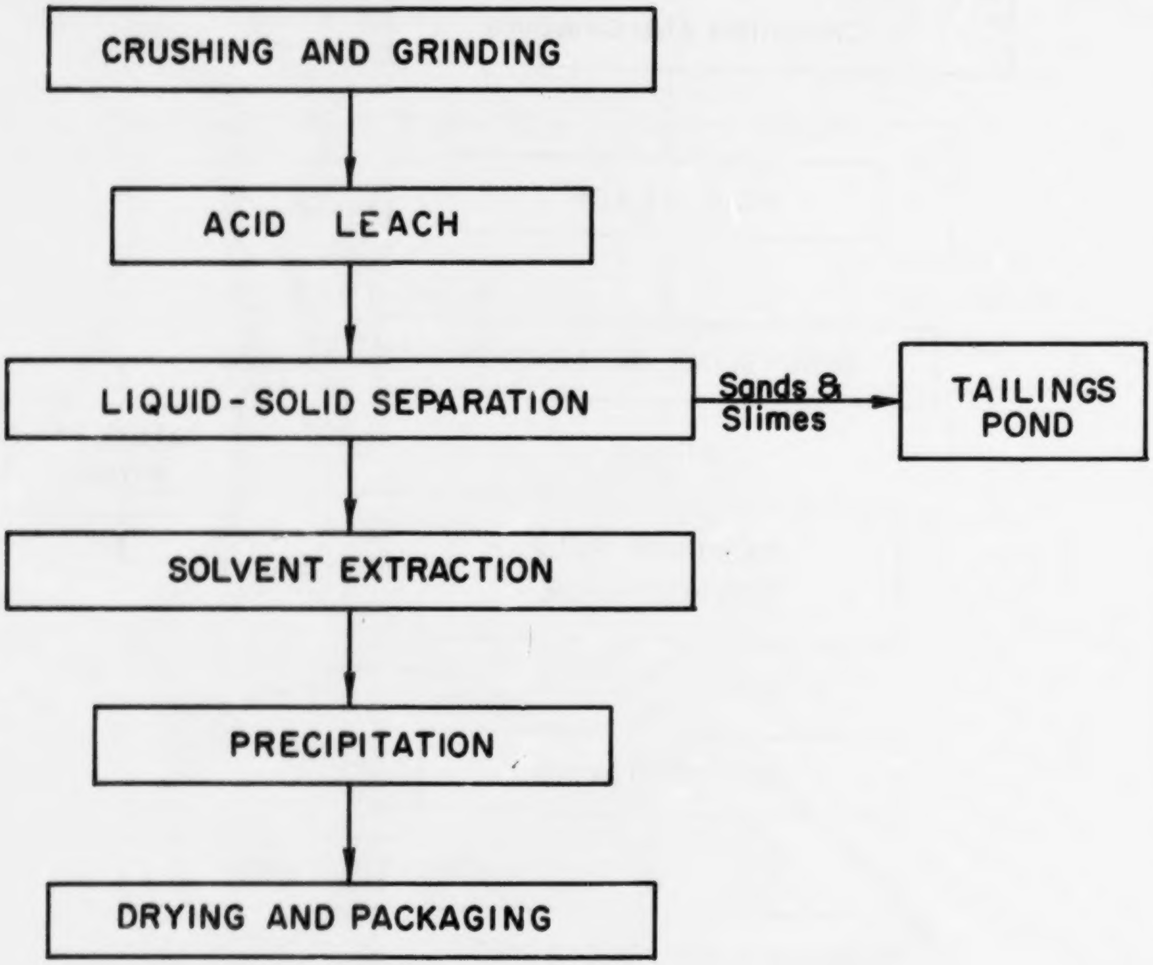

CRUSHING AND GRINDING
ACID LEACH
LIQUID-SOLID SEPARATION
Sands &
Slimes
TAILINGS
POND
SOLVENT EXTRACTION
PRECIPITATION
DRYING AND PACKAGING

Figure 3. PROCESS FLOW DIAGRAM A

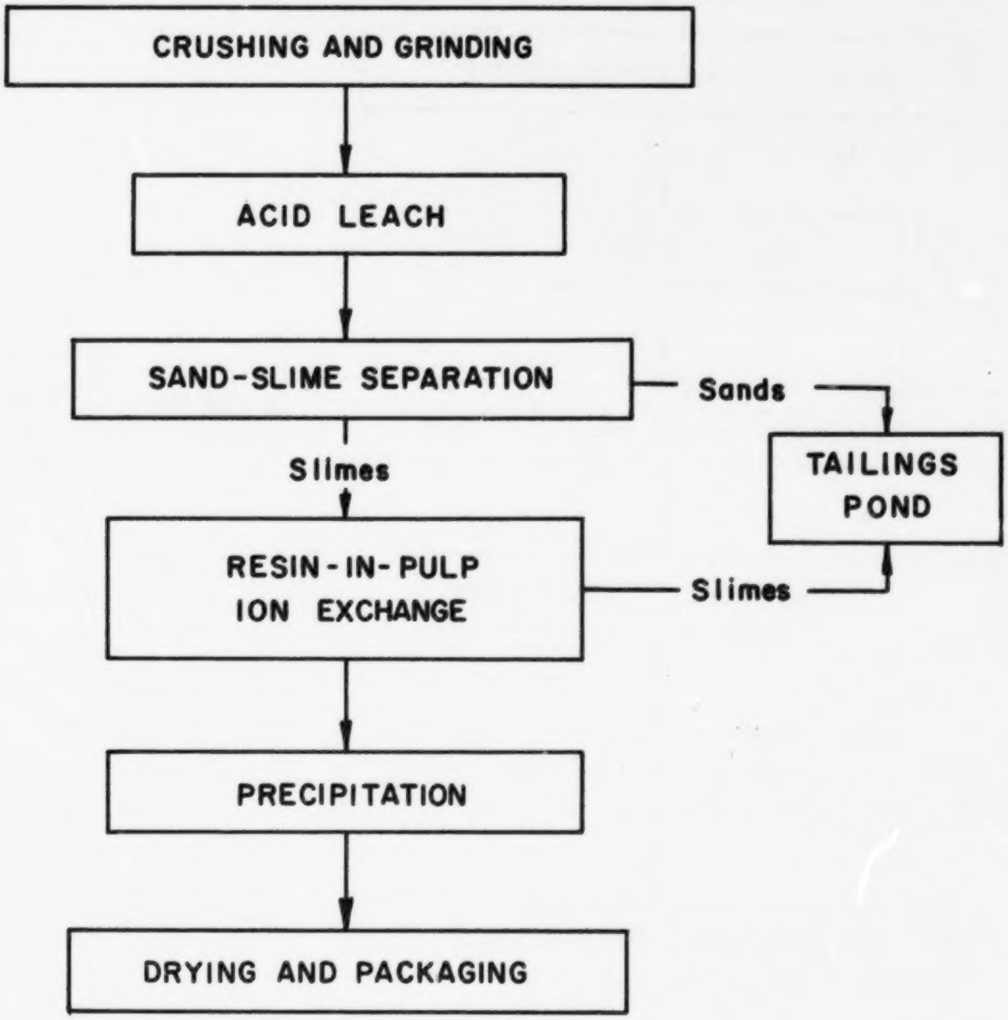

CRUSHING AND GRINDING
ACID LEACH
SAND-SLIME SEPARATION
Sands
TAILINGS POND
Slimes
RESIN-IN-PULP ION EXCHANGE
Slimes
PRECIPITATION
DRYING AND PACKAGING

Figure 4. PROCESS FLOW DIAGRAM B

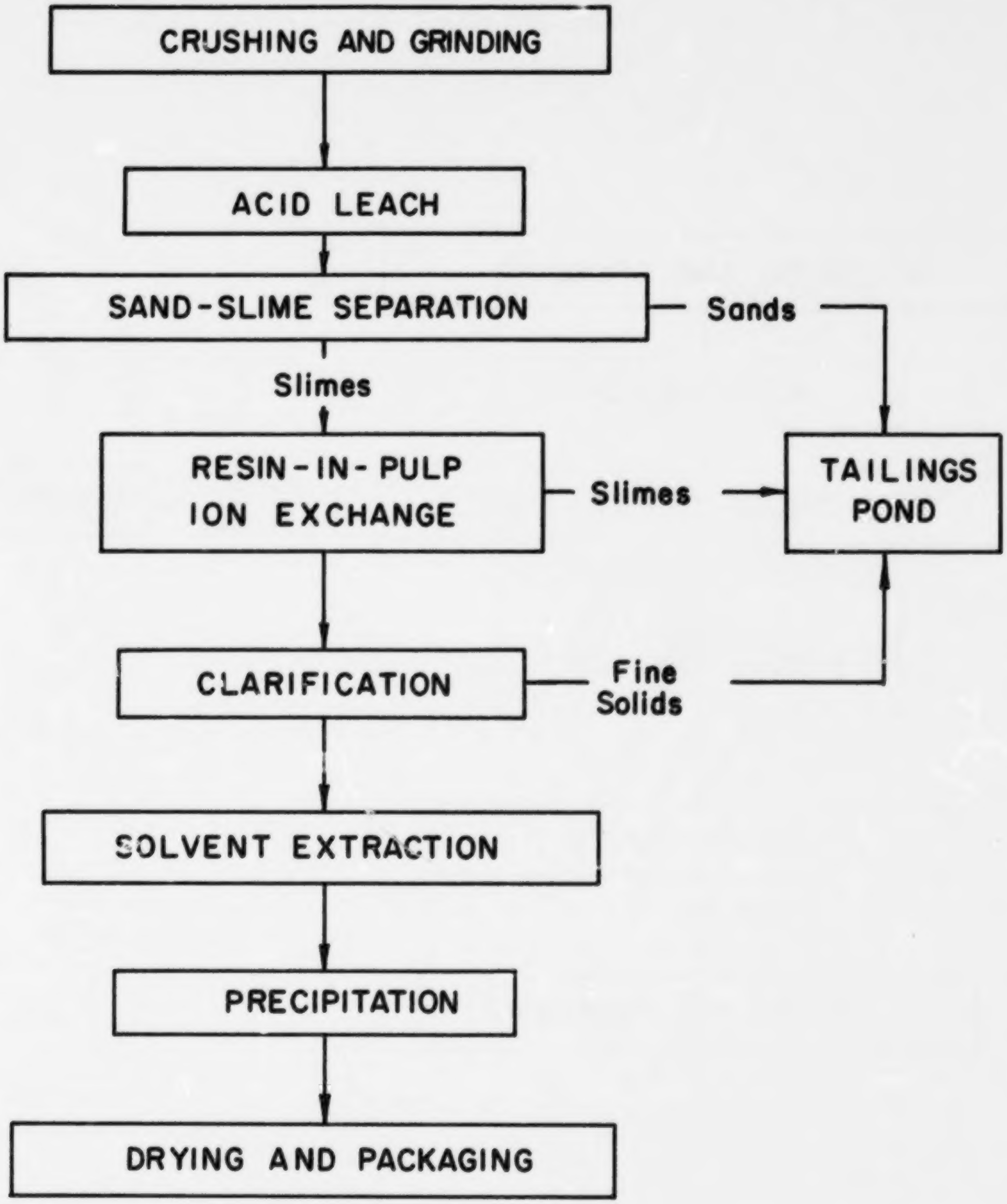

CRUSHING AND GRINDING
ACID LEACH
SAND-SLIME SEPARATION
Sands
Slimes
RESIN-IN-PULP ION EXCHANGE
Slimes
TAILINGS POND
CLARIFICATION
Fine Solids
SOLVENT EXTRACTION
PRECIPITATION
DRYING AND PACKAGING

Figure 5. PROCESS FLOW DIAGRAM C

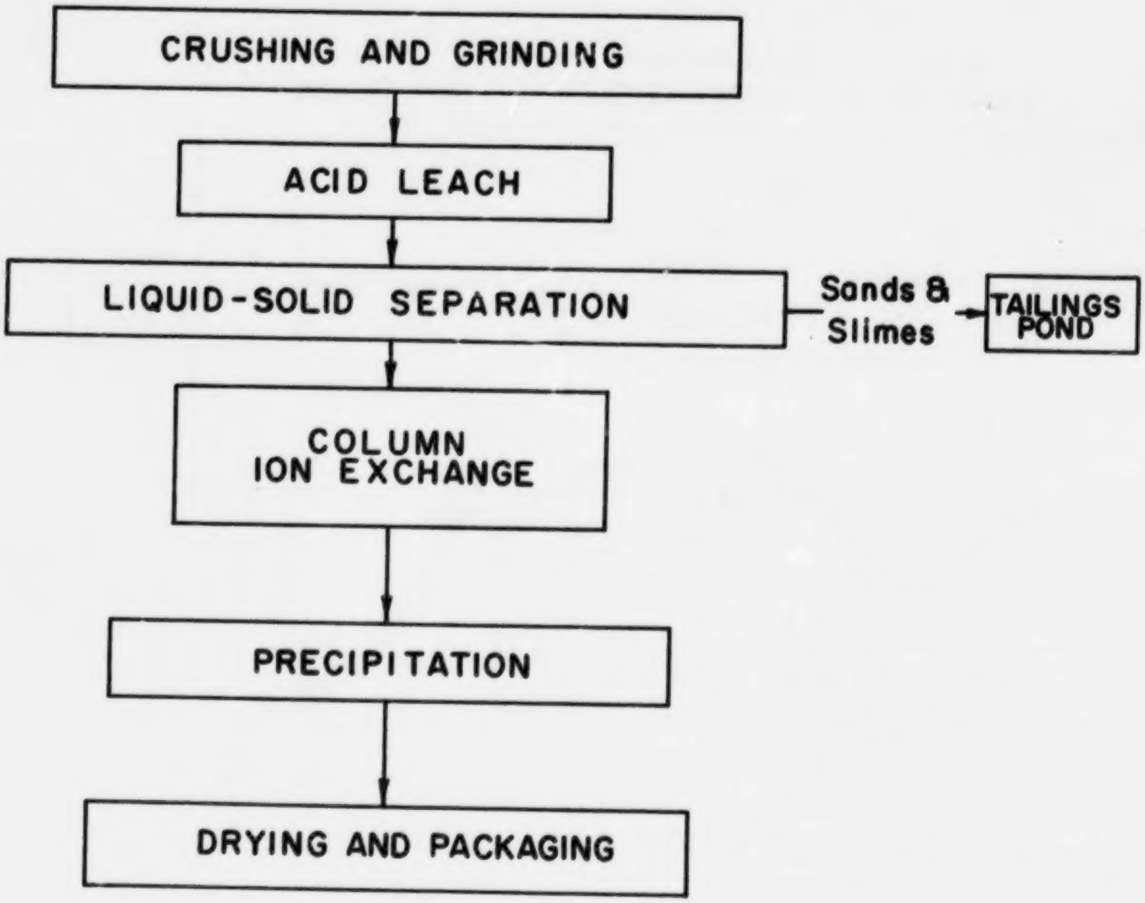

CRUSHING AND GRINDING
ACID LEACH
LIQUID-SOLID SEPARATION
Sands & Slimes
TAILINGS POND
COLUMN ION EXCHANGE
PRECIPITATION
DRYING AND PACKAGING

Figure 6. PROCESS FLOW DIAGRAM D

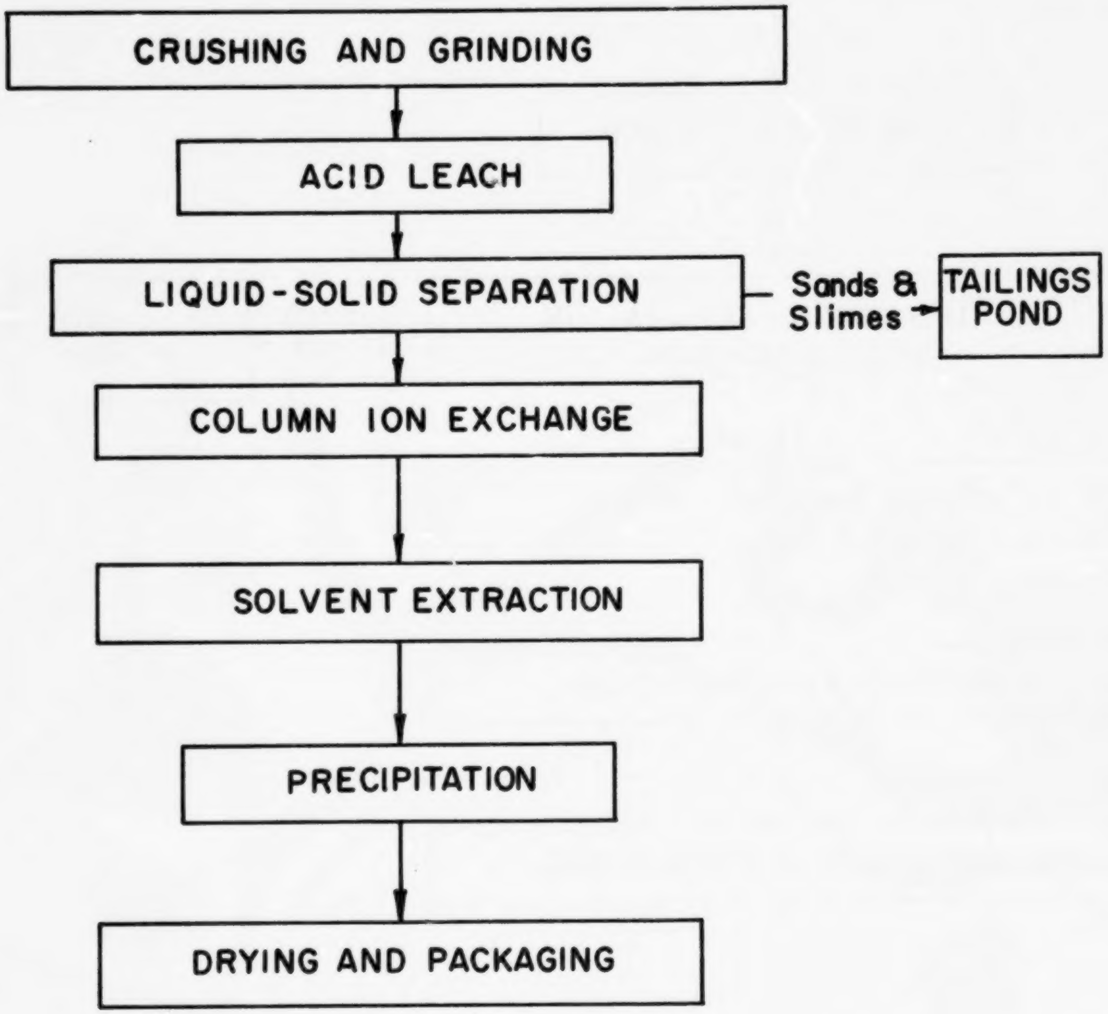

CRUSHING AND GRINDING
ACID LEACH
LIQUID-SOLID SEPARATION
Sands & Slimes
TAILINGS POND
COLUMN ION EXCHANGE
SOLVENT EXTRACTION
PRECIPITATION
DRYING AND PACKAGING

Figure 7. PROCESS FLOW DIAGRAM E

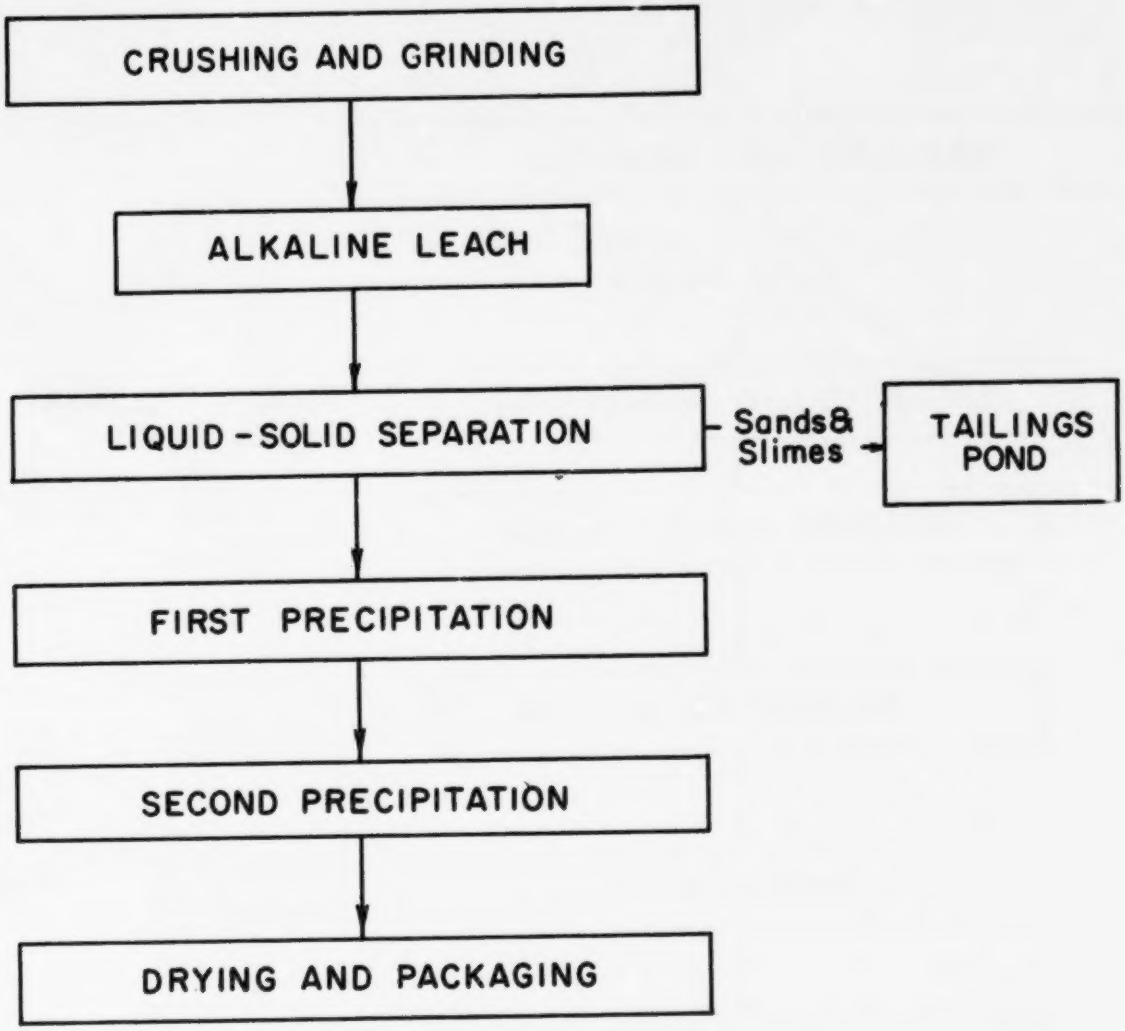

CRUSHING AND GRINDING
ALKALINE LEACH
LIQUID - SOLID SEPARATION
Sands & Slimes
TAILINGS POND
FIRST PRECIPITATION
SECOND PRECIPITATION
DRYING AND PACKAGING

Figure 8. PROCESS FLOW DIAGRAM F

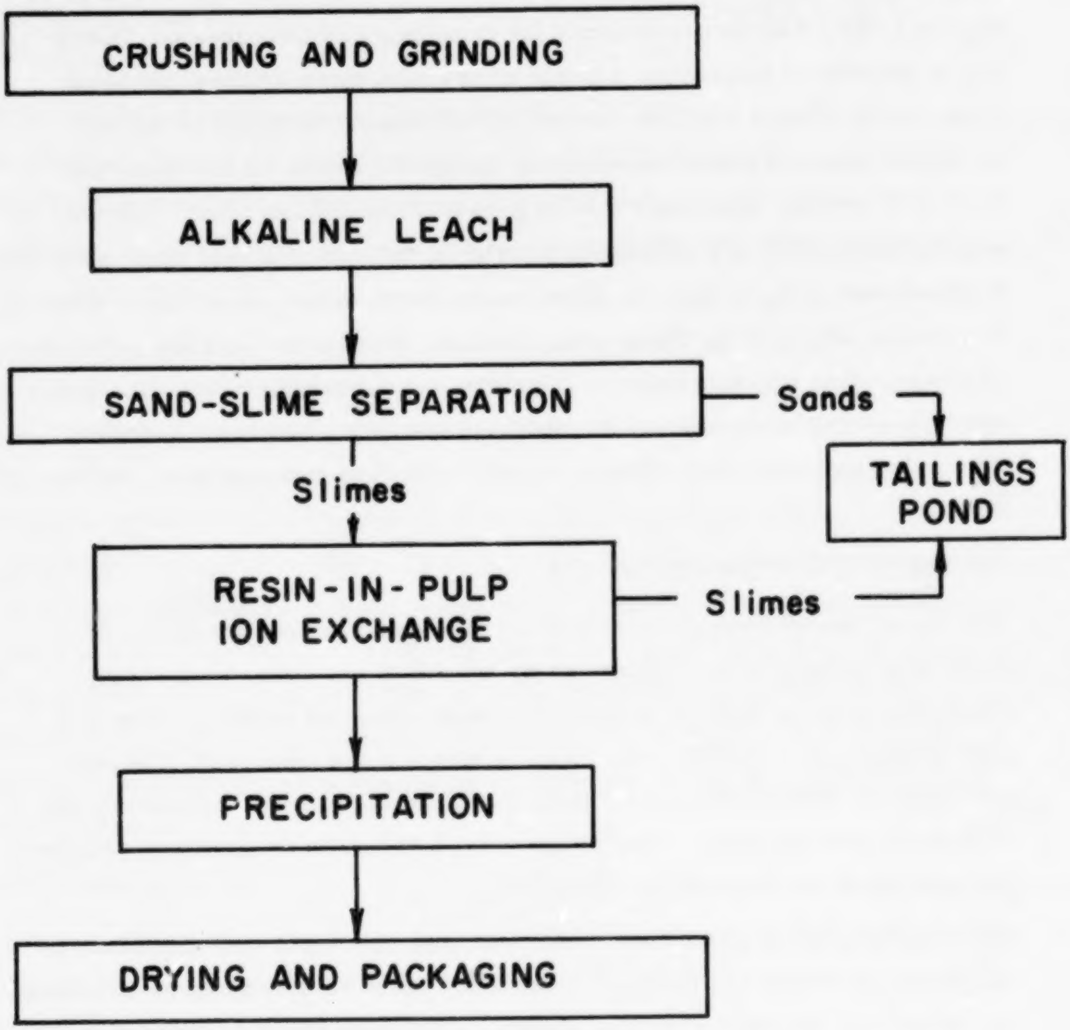

CRUSHING AND GRINDING
ALKALINE LEACH
SAND-SLIME SEPARATION
Sands
TAILINGS POND
Slimes
RESIN-IN-PULP ION EXCHANGE
Slimes
PRECIPITATION
DRYING AND PACKAGING

Figure 9. PROCESS FLOW DIAGRAM G

ORE HANDLING

The majority of operating mills in the United States blend the ore to some degree before delivery to the mill for crushing and processing. Blending is performed to provide a grade of ore with more uniform physical characteristics and uranium content. Radiometric analysis of the ore as loaded onto the trucks establishes the grade which varies from less than 0.10 percent uranium oxide to greater than 1.0 percent. The ore may be stockpiled in a manner to provide a uniform grade of approximately 0.25 percent to the crusher. Ores can be hard, slimy, or sandy, which may cause difficulty in the milling circuits. Hard ores limit the capacity of the grinding circuits, slimes interfere in ion exchange circuits, and sandy ores settle too rapidly in pipelines causing plugging. Blending eliminates extremes and allows a smoothly flowing process to be maintained.

CRUSHING AND GRINDING OF ORE

The ore is moved from the stockpiles to the crusher feed by means of front-end loaders or bulldozers or by trucks to receiving bins. Jaw crushers ranging from 15 to 40 inch size are used as primary units but may be bypassed for fine ore. Grizzly circuits are employed to screen and remove undersize material from the crushing circuit, bypassing the fine ore to storage bins. Both impact-type and cone or gyratory crushers are also used as required by ore type.

Moisture content is important in the crushing operation and should be in the range of five to ten percent. Some ore must be dried before crushing by either kiln drying or natural drying. After crushing to sizes of approximately 1/4 to 1 inch, the ore is carried by a conveyer belt to the fine ore bins where it is held until needed for processing.

Some mills employ a roasting circuit for use with special types of ore. Pretreatment of lignite ores by roasting improves the leaching characteristics. Improved settling and filtration characteristics of clay minerals are obtained by roasting. Vanadium-bearing ores require a salt-roasting process to improve the solubility of vanadium. Carbonaceous ores are roasted to remove organic carbon and prevent contamination of leach solutions.

The roasting circuit consists of feeding the ore from the crushing circuit through a rotary kiln at a temperature of 600° F and returning the ore to the grinding circuit. The roasting circuit is bypassed when not needed with the ore going directly from the crushing to the grinding circuit.

The ore is carried by belt-type feeders at the desired feed rate to the grinding circuit and sampled at some point between the crushing and grinding circuit for laboratory analysis. Rod mills and ball mills are used almost exclusively for grinding the ore to approximately 28 mesh for an acid leach process and 200 mesh for an alkaline leach process. Water is added to obtain a slurry of approximately 65 percent solids for grinding. Recycled acidic wash solutions from other plant circuits are sometimes added to reduce the water requirements. Classifiers are sometimes placed in a closed circuit with the grinding equipment to size the ore and return coarser particles for further grinding. In some instances, the pulp density in the grinding circuit is different from that required for the leaching circuit and must be adjusted by means of cyclones and thickeners.

ACID LEACH

The leaching process selected for removal of uranium from the ore is dependent on the physical and chemical characteristics of the ore. Among these are the type of uranium mineralization, ease of liberation, and the nature of other constituent minerals present. The most important factor in choosing the leaching process, however, is based on the lime content of the ore. Ores with low lime content (12% or less) are leached with acid, while those with a high lime content require large quantities of acid for neutralization purposes; hence, employ the alkaline leach process for economic considerations. Sulfuric acid, rather than hydrochloric or nitric, is commonly utilized for leaching purposes due to its less corrosive nature and lower cost.

Uranium in the ore in the tetravalent form must be oxidized to the hexavalent state before dissolution occurs. Iron present in the ore or introduced from wear of the metal in the grinding circuit serves as the principle oxidant. To be effective, however, the iron must be present in the oxidized or ferric state. Sodium chlorate or manganese dioxide is employed for this purpose. The oxidation-reduction potential (emf) of the ore slurry is important and should lie within the range of -400 to -500 millivolts. Failure to maintain uranium in the hexavalent state throughout the leaching process will result in premature precipitation.

The ore slurry is pumped into the leach circuit maintaining a pulp density of 50 percent solids ground less than 28 mesh. Leaching is performed in a series of wooden tanks equipped with agitators. Sulfuric acid is added in an amount to maintain a free acid concentration ranging from 1 to 90 grams acid per liter (40 to 120 pounds per ton). Higher acid concentrations are used for vanadium extraction due to its greater insolubility.

CARBONATE LEACH

As mentioned previously, the alkaline sodium carbonate leaching process is utilized for ores containing excessive amounts of lime. Since many of the minerals in the ore do not react with the carbonate solution, the ore must be ground much finer so as to expose more uranium mineral surface area for efficient leaching. The ore is normally ground to 70 to 80 percent less than 200 mesh size.[4] Factors that affect the leaching rate of uranium are temperature, pressure, contact time, oxidation, reagent concentration, degree of agitation, fineness of grind, and pulp density. Due to the selectivity of the carbonate leach solution, uranium may be precipitated without further treatment following liquid-solid separation.

The carbonate leaching process consists of the transfer of a 65 percent slurry from the grinding circuit to a thickener circuit that adjusts the slurry to about 55 percent solids. Carbonate solution from the precipitation circuit is adjusted for chemical content and recycled to the grinding circuit to make the slurry.

The adjusted slurry is pumped to a series of Pachuca leach tanks and heated to 180° F with steam. The leach solution contains 40 to 50 grams of sodium carbonate per liter and 10 to 20 grams of sodium bicarbonate per liter. Bicarbonate is added to prevent reprecipitation of the dissolved uranium through reaction with the hydroxyl ion. Air is bubbled through the solution to oxidize uranium from the tetravalent to the hexavalent state. The circular tanks are 20 feet in diameter and 40 to 60 feet deep; pressure from the solution depth increases the leaching rate. Leaching time varies from 24 to 72 hours depending on the ore characteristics and plant operating conditions. Pressure leaching prior to or following the atmospheric leaching process is used to increase the rate of leaching in some milling circuits.

LIQUID-SOLID SEPARATION

After the ore has been processed through the acid leach or carbonate leach circuit, the loaded leach liquid must be separated from the sands and slimes prior to entering ion exchange or solvent extraction circuits. The resin-in-pulp ion exchange process represents an exception to the separation procedure in that the slimes do not have to be removed from the leach slurry.

Separations are accomplished by countercurrent decantation washing in cyclones, classifiers, and thickeners, or by filtration. Countercurrent decantation washing procedures are the most efficient and current means of separating solids from acid leach liquid. The method is used in all but one of the acid leach process mills with washing efficiencies better than 99 percent. The circuit usually has five or more stages of countercurrent washing in which the wash solution and solids flow in opposite directions. Flocculants are added to increase separation rates. Pressure precoat filters or activated carbon are used to further clarify the feed solution for the Amex solvent extraction process.

Complete separation of liquid and solids is unnecessary for resin-in-pulp ion exchange processes. Cyclones and classifiers are used to separate sands from slimes of less than 325 mesh size. Sands are wasted to the tailings pond.

Alkaline leach liquids and solids are separated by vacuum drum filtration to minimize the volume of wash water required. The solids are repulped and wasted to the tailings pond.

ION EXCHANGE

Ion exchange circuits are employed to concentrate and purify the leached uranium. Strong and intermediate base anionic type resins are loaded from either a sulfuric acid or a carbonate leach feed solution. The loaded resin is stripped with a chloride, nitrate, bicarbonate, or an ammonium sulfate-sulfuric acid solution.

Four types of ion exchange circuits are utilized by the processing mills. The fixed-bed type consists of stationary columns packed with resin. As the feed solution passes through, uranium is sorbed on the resin. The resin is washed and the uranium desorbed. The extraction columns are sensitive to plugging by solids in the feed solution. The moving-bed column circuit has stationary columns, but resin is transferred to different columns to perform loading, washing, and eluting operations. In the continuous resin-in-pulp process, sorption, washing, and desorption are performed by contacting the resin and process solutions in a series of tanks. The resin and solution flow countercurrently in the tanks and are separated by screens. Forced air is employed for agitation. The basket resin-in-pulp process utilizes a series of resin-filled cubical baskets that are jigged up and down in the process solution contained

ELUEX PROCESS

The Eluex process is a combination ion exchange-solvent extraction process.[4] The eluate produced by sulfuric acid elution of the ion exchange resin is fed to either the Dapex or Amex solvent extraction process. The Eluex process eliminates the requirement for nitrate and chloride reagents, thus preventing potential pollution by these elements. Additionally, a purer end product is obtained.

PRECIPITATION

Uranium is precipitated from solution by addition of sodium hydroxide, gaseous ammonia, hydrogen peroxide, or magnesia. Several stages of precipitation at controlled pH are often used with the pH being readjusted in a precipitation tank near the end of the circuit. Gaseous ammonia is used as the precipitating agent in most mills. When sodium hydroxide is used to precipitate uranium, resolution with sulfuric acid followed by reprecipitation with gaseous ammonia produces a purer product with little sodium remaining.

DRYING AND PACKAGING

The slurry from the precipitation circuit is dewatered in thickeners followed by drum, plate, or frame filters. The filtercake is repulped and transferred to a multiple hearth dryer operating at 700° to 800° F. The dried product containing approximately 96 percent uranium oxide is crushed and sealed in shipping drums.

SECONDARY METAL RECOVERY

In several instances, other metals are present in uranium ores in sufficiently high concentrations to warrant economic extraction. The Atlas Corporation mill at Moab, Utah, extracts copper as a by-product by means of a flotation circuit. Copper, cobalt, and nickel are recovered from the acid leach process solutions by the Cotter Corporation at Canon City, Colorado. Vanadium is extracted at two Union Carbide mills at

Uravan and Rifle, Colorado, and also by the Mines Development Corporation mill in South Dakota. Molybdenum is present in some ores in concentrations sufficient to cause fouling of the extraction circuits in the mills and must be stripped. Concentrations of molybdenum in ores mined in Texas are sufficient to justify by-product recovery.

SECTION VII

REFINING PROCESSES

Uranium concentrate must be further purified and converted to uranium hexafluoride before utilization as a feed for gaseous diffusion plants. The process is performed in government-owned operations and two commercially-owned refineries, the Allied Chemical Corporation plant on the Ohio River at Metropolis, Illinois, and the Kerr-McGee Corporation facility on the Illinois River at Gore, Oklahoma. The commercially-operated plants will be discussed below.

In the Kerr-McGee process the uranium concentrate feed is digested with hot nitric acid to solubilize uranium and countercurrently extracted with tributyl phosphate in hexane.[24] The loaded organic is then stripped of uranium with water, concentrated in a two-stage heating process, dehydrated and denitrated in a stirred reactor to form pure uranium trioxide. Next, uranium trioxide is fed to a two-stage fluid bed operation at approximately 1100° F, and reduced to uranium dioxide by countercurrent flow of dissociated ammonia. Conversion to uranium tetrafluoride is accomplished by countercurrent contact with a stream of anhydrous hydrogen fluoride gas. Final conversion to uranium hexafluoride is by contact with fluorine gas in a series of fluorination towers. The gaseous product is cooled and filtered twice by sintered metal particulate filters, condensed to a solid at 50° F in a cold trap, and sealed in 10-ton cylinders for shipping. The Gore, Oklahoma, facility is presently processing approximately 5,000 tons per year of uranium concentrate, but can be expanded to 10,000 tons per year.

The primary waste from the refining process is the raffinate from the solvent extraction circuit. The solution contains approximately one molar concentration of nitric acid, nitrates, radium-226, and other radioactive impurities. The waste is permanently retained in an evaporation pond after neutralization with lime. Injection well waste disposal is planned to replace evaporation ponds, if approval can be obtained.

The waste solution from the fluorination process, containing 0.3 percent hydrofluoric acid, is treated with calcium fluoride. Sulfuric acid is then added to precipitate excess calcium and to neutralize the effluent before discharge.

Large quantities of cooling water are used in the plant and returned to the river with little change in temperature. Contaminated air is passed through vacuum transfer and cleaning systems prior to atmospheric discharge.

The Allied Chemical process eliminates the acid dissolution and solvent extraction purification steps.[11] The uranium concentrate is reacted in a series of fluid beds, reduced to uranium dioxide, and converted to crude uranium hexafluoride. The crude product is purified by fractional distillation. Raffinate wastes are eliminated in the process since a solvent extraction circuit is not used. Uranium concentrate impurities are present in purification circuit wastes.

A refinery produces liquid wastes containing 5.2 grams radium-226 per kiloton of processed alkaline leach concentrate and 0.26 grams per kiloton of processed acid leach concentrate. In comparison, a mill produces liquid waste containing 0.1 grams of radium-226 per kiloton of alkaline leach concentrate and 0.77 grams per kiloton of acid leach concentrate. This estimate demonstrates that dissolved radium-226 in refinery wastes can be greater than in uranium mill wastes.

Small losses of uranium in liquid wastes and air may occur but should be far below the allowable maximum permissible concentration. Although the concentration is low, uranium will be the chief contributor to the gross alpha activity of the waste and will mask alpha activity of radium-226.

SECTION VIII

WASTE CHARACTERISTICS

In-plant surveys were conducted in uranium mills representing all of the typical processing methods in 1962.[25] Findings from the studies, as well as others, were used in the assembly of a waste guide manual for the uranium industry.[26] While milling processes have changed to some extent through the development of new technology, the processes used today are basically similar to those in use at the time of the 1962 surveys. The waste characteristics will be discussed below according to types of waste products.

MILL PROCESS EFFLUENTS

During processing, one to three percent of the ore is dissolved in the mill process water. Other chemicals used in the milling process contribute to the dissolved solids content of the effluent. Concentrations vary according to the leaching process utilized. During leaching of the ore with sulfuric acid solutions, a large portion of the acid is consumed in chemical reactions and cannot be reused for extraction purposes. Additionally, large volumes of water are utilized in countercurrent decantation washing of the leached ore solids. The spent acid and wash waters are discharged to tailings ponds in volumes of approximately 1,000 gallons per ton of ore processed.

The alkaline leach process utilizes a sodium carbonate-sodium bicarbonate leaching solution to extract uranium from the ore solids. The solution is not consumed in the process and is reusable following separation of the uranium and recarbonation. Ore solids are separated from the leach liquor by a filtration process rather than washing, thus reducing water requirements to 250 gallons per ton of ore processed. To prevent a buildup of dissolved solids, a portion of the alkaline leach process water is discharged to the tailings pond and fresh water added to replenish the volume. Tailings pond water is returned to the mill to reslurry sands and slimes for disposal.

Other milling process solutions from the precipitation, solvent exchange, and ion exchange circuits are partially recovered and recycled.

SANDS AND SLIMES

The spent ore solids are discharged to the tailings pond as a slurry by mixing with water or mill process effluents. Approximately 97 percent of the ore processed through the mill leaching circuit remains in a solid form and is discharged from the mill to the tailings pond as spent ore solids--80 percent sands and 20 percent slimes.

SUSPENDED SOLIDS IN MILL EFFLUENTS

The greater majority of solid particles settle to the bottom of the pond leaving an apparently clear liquid. However, a small amount of fine ore particles are held suspended in the mill effluent or are resuspended from settled solids through agitation of the liquid. The weight of these solids varies from 10 to 500 milligrams per liter.

MINE WATER DRAINAGE

Mining operations for uranium are being performed at greater depths as ore near the surface is being depleted. At the deeper depths, ground water often flows through the ore bodies and dissolves chemical constituents from the ore. The water must be pumped from the mining area to permit removal of the ore. Mine water is discharged from mines in the Ambrosia Lake area of New Mexico at a rate of five million gallons per day.[27] Open pit mines in the Gulf Coast Area of Texas release 840,000 gallons per day. The mine water is usually discharged to the surface to flow into a nearby waterway or seep into the ground. Mine water from an operating mine is pumped into a nearby abandoned mine by a company in the Texas Gulf Coast Area. Uranium is recovered from mine water by ion exchange methods at several locations prior to discharge.

AIRBORNE RADIOACTIVITY

Airborne radioactivity in the form of a gas or fine solid particles is found in uranium mines, in areas of stockpiled ore awaiting processing, and in waste disposal areas. Radioactive dust is found in mill areas concerned with crushing and grinding, precipitation, and packaging. In the surrounding vicinity of the mill, airborne radioactivity may be created by mining operations, or by wind erosion of the stockpiled ore or the dry tailings solids.

The radioactive gas emitted from the ore is found in the greatest concentrations in restricted areas of poor ventilation, such as underground mines. The gas will also accumulate around stockpiled ore and tailings areas under conditions of limited air movement.

CHEMICAL CLASSIFICATION OF WASTES

INORGANIC POLLUTANTS

The sources of inorganic pollutants in mill discharges are chemicals utilized in the milling process and solubilized materials from the ore. Inorganic chemicals employed in both the acid and alkaline leach processes are listed in Table 16. Metallic elements that may be solubilized during the milling process are enumerated in Table 17. Some mills economically recover a number of these elements, thus reducing a potential source of pollution. Chlorides, nitrates, and sulfates are also solubilized during the milling process.

The milling process chemicals are somewhat different from those used several years ago.[26] Ammonium sulfate has replaced ammonium nitrate in most current milling processes. Air is used as the oxidizing agent in the alkaline leach process rather than chemicals. The majority of the mills employ ammonia for precipitation of the uranium.

ORGANIC POLLUTANTS

Organic chemicals are introduced into the processing circuit during liquid-solid separation and solvent extraction. The chemicals used are listed in Table 16. Organic extractants are recycled in the milling process. Due to incomplete phase separation, however, the organic loss is approximately one-half gallon per 1,000 gallons of solution passing through the solvent extraction circuit.

RADIOACTIVE POLLUTANTS

The average grade of uranium-bearing ore is 0.22 percent uranium oxide.[28] The uranium is present as uranium-238 and uranium-234 from the uranium-radium family and uranium-235 from the uranium-actinium family. Natural uranium contains 99.28 percent uranium-238, 0.0057

Table 16. CHEMICALS USED IN MILLING OPERATIONS

Acid leach process	Alkaline leach process
Acid Leach Circuit: sulfuric acid sodium chlorate	Alkaline Leach Circuit: sodium carbonate sodium bicarbonate
Liquid-Solid Separation Circuit: polyacrylamides guar gums animal glues	Ion-Exchange Circuit: strong base anionic resins sodium chloride sulfuric acid sodium bicarbonate ammonium nitrate
Ion-Exchange Circuit: strong base anionic resins sodium chloride sulfuric acid sodium bicarbonate ammonium nitrate	Precipitation Circuit: ammonia gas magnesium oxide hydrogen peroxide
Solvent Extraction Circuit: tertiary amines (usually alamine-336) alkyl phosphoric acid (usually EHPA) isodecanol tributyl phosphate kerosene sodium carbonate ammonium sulfate sodium chloride ammonia gas hydrochloric acid	
Precipitation Circuit: ammonia gas magnesium oxide hydrogen peroxide	

Table 17. METALS LEACHED FROM ORE BY MILLING PROCESS[4, 26]

Magnesium
Copper
Manganese
Barium
Chromium
Molybdenum
Selenium
Lead
Arsenic
Vanadium
Iron
Cobalt
Nickel

percent uranium-234, and 0.71 percent uranium-235. The uranium-radium family is of primary concern as a radioactive waste since it is the major constituent of natural uranium in the ore.

Uranium-238 is the first member of a long series of radioactive isotopes which decay to stable lead-206. The series contain eight alpha emitters and six beta emitters. The decay chain for the radioactive family is shown in Figure 10. Two minor branches occur but are not shown because their effect is negligible. As seen in Figure 10, uranium-238 decays by an alpha emission to thorium-234 with a half-life of 4.5 billion years; the thorium-234 decays by beta emission to protactinium-234 with a half-life of 24.1 days; the protactinium-234 decays by beta emission to uranium-234 with a half-life of 1.1 minutes with decay continuing until stable lead-206 is formed. Most ores occur with the members of the radioactive family in equilibrium, the state that prevails when the ratios between the amounts of successive members of family remain

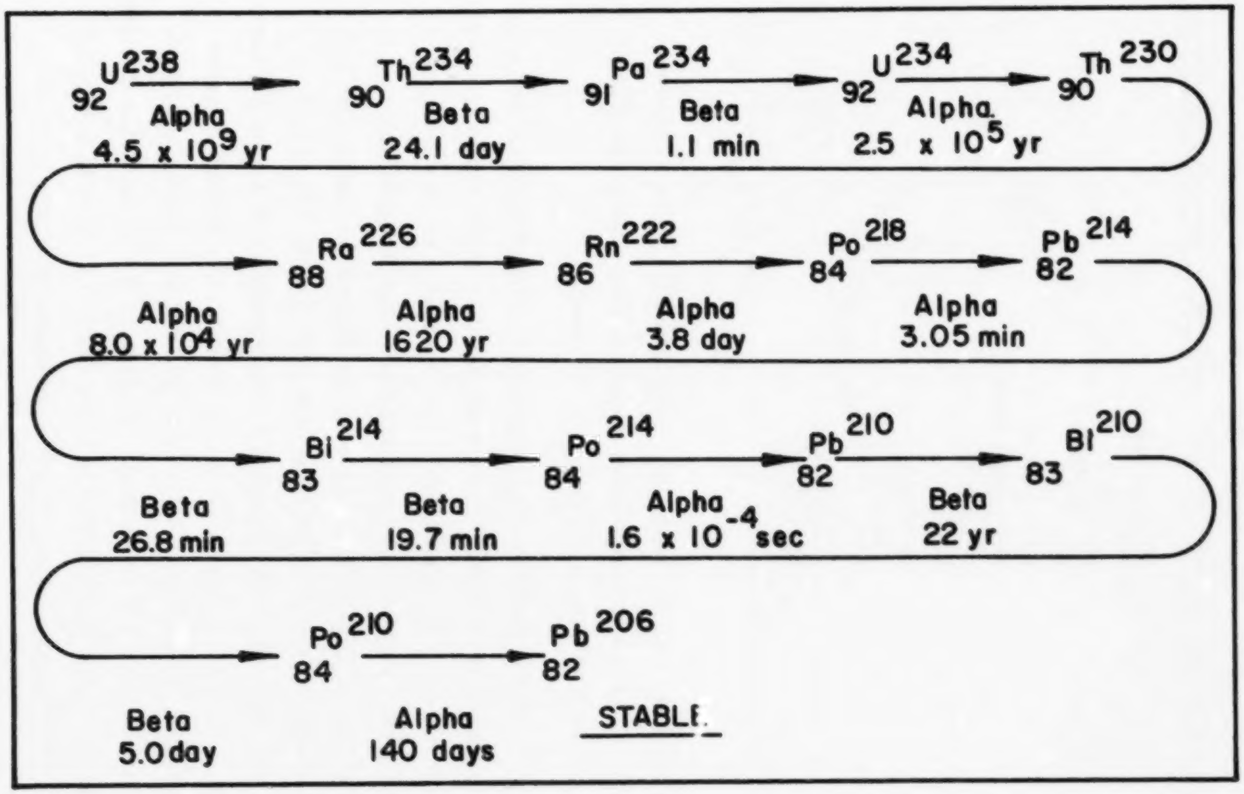

$_{92}U^{238}$
Alpha
4.5 x 10^9 yr
$_{90}Th^{234}$
Beta
24.1 day
$_{91}Pa^{234}$
Beta
1.1 min
$_{92}U^{234}$
Alpha
2.5 x 10^5 yr
$_{90}Th^{230}$
Alpha
8.0 x 10^4 yr
$_{88}Ra^{226}$
Alpha
1620 yr
$_{86}Rn^{222}$
Alpha
3.8 day
$_{84}Po^{218}$
Alpha
3.05 min
$_{82}Pb^{214}$
Beta
26.8 min
$_{83}Bi^{214}$
Beta
19.7 min
$_{84}Po^{214}$
Alpha
1.6 x 10^{-4} sec
$_{82}Pb^{210}$
Beta
22 yr
$_{83}Bi^{210}$
Beta
5.0 day
$_{84}Po^{210}$
Alpha
140 days
$_{82}Pb^{206}$
STABLE

Figure 10. URANIUM-RADIUM FAMILY (minor branches not shown)

constant. One million years are required to obtain the equilibrium. Natural leaching by ground water of some members of the family may disturb the equilibrium and has occurred in some ores.

Uranium-238 has an alpha activity of 152 microcuries per pound. If the series is in equilibrium, each of the daughters will have the same activity. The eight alpha emitting daughters will have an activity of 1,216 microcuries per pound of uranium and the six beta emitting daughters will have an activity of 912 microcuries per pound, resulting in a total alpha and beta activity of 2,128 microcuries per pound of uranium. As only uranium is recovered from the ore, the other radioactive members of the family are discharged as waste. The activity of the recovered alpha-emitting uranium isotopes is 304 microcuries per pound, resulting in the discharge as mill waste of the other 12 alpha and beta-emitting isotopes with an activity of 1,824 microcuries per pound; hence, 85 percent of the total activity is contained in the mill waste.

Because of the importance of radium-226 as a pollutant, a detailed survey was made with one of the main objectives to determine the radium distribution throughout the milling process.[25] A study of an acid leach process revealed that 0.2 to 0.4 percent of the radium contained in the ore was dissolved; 0.05 to 0.10 percent was precipitated along with the uranium concentrate, and 0.15 to 0.30 percent was discharged to the tailings pond in the liquid waste from solvent extraction or ion exchange circuits. The remaining 99.6 to 99.8 percent of the radium was discharged into the tailings pond in sands and slimes. It was also determined that the radium-226 distribution between the sands and slimes was 20 and 80 percent, respectively.

A greater amount of radium is dissolved from the ore in an alkaline leach process. From the study it was found that 1.5 to 2.0 percent of the radium was dissolved and precipitated with the uranium concentrate. The remaining 98.0 to 98.5 percent was discharged into the tailings pond in the sands and slimes. Although none of the mill surveys were performed at an alkaline leach mill using an ion exchange circuit, it is probable that the majority of the dissolved radium would be separated from the uranium at this point, thus producing a uranium concentrate with much less radium present.

Using 0.22 percent as the average grade of uranium oxide in ore, the radium-226 concentration in the ore is 0.6 milligrams per ton of ore processed. The uranium ore processing plants operating in 1971, with a maximum production capacity of 27,650 tons per day, had a discharge potential of 16.0 grams of radium per day in the mill waste.

Since uranium is the radionuclide being recovered, only small quantities will be present in the waste material. An extraction circuit is being installed in one of the Texas mills to recover the small amount of uranium discharged.

The distribution of the other members of the uranium-radium family in the mill waste streams has not been investigated to the extent of radium-226. Laboratory analyses were performed on mill effluents from mills located in the Colorado River basin for several years by the Colorado River Basin Project Laboratory located in Salt Lake City, Utah. Results of analyses in Tables 18 through 22 illustrate the levels of concentrations typical of uranium mill effluents. The effluents were processed through waste treatment circuits for radium-226 removal prior to laboratory analysis.

Table 23 shows concentrations of radium-226 in mill effluent suspended solids. The concentration is also expressed as picocuries of radium in suspended solids contained in one liter of effluent. The radium content from suspended material is, in general, significantly greater than the dissolved radium content after waste treatment.

Radon-222, the daughter of radium-226, is a radioactive gas and may be found in poorly ventilated areas. The isotope is of greatest concern in underground mining operations. It may be seen from Figure 10 that radon-222 has four, short, half-life daughters: polonium-218, lead-214, bismuth-214, and polonium-214. The daughters of radon-222 are solids, but when formed in air, attach quickly to any solid surface, such as dust particles. In this manner the radionuclides may remain suspended for prolonged periods.

Table 18. MILL EFFLUENT NO. 1

Sample No.	Ra-226 pc/l	U µg/l	Pb-210 pc/l	Po-210 pc/l	Th (alpha) pc/l	Gross alpha pc/l	Gross beta pc/l
1	8.8	830	1,020	707.0	54.0	1,100	2,100
2	21.2	160	660	606.0	54.0	700	1,400
3	13.0	220	396	232.0	90.0	340	1,000
4	23.2	500	920	198.0	117.0	400	1,700
5	10.8	88	207	135.0	19.0	230	810
6	7.5	50	120	19.0	16.0	71	690
7	12.0	400	250	4.4	20.0	480	1,100
8	10.0	680	552	85.0	5.2	860	1,400
9	13.0	490	790	99.0	9.4	700	1,500
10	21.0	1,800	910	70.0	300.0	2,000	2,900
11	8.4	1,400	430	210.0	120.0	2,200	2,000
12	13.0	740	500	336.0	21.0	760	1,700
13	14.0	510	840	340.0	17.0	600	2,000
14	17.0	410	1,250	-	10.0	800	2,700
15	14.0	360	1,090	990.0	24.0	940	2,400
16	8.5	140	460	77.0	13.0	380	1,200
17	10.8	32	2	2.2	3.3	88	780
18	18.7	510	773	528.0	21.0	1,100	2,300

Table 19. MILL EFFLUENT NO. 2

Sample No.	Ra-226 pc/l	U µg/l	Pb-210 pc/l	Po-210 pc/l	Th (alpha) pc/l	Gross alpha pc/l	Gross beta pc/l
1	29	3,500	6.2	2.6	364.0	2,100	4,200
2	15	3,800	5.0	4.5	3,680.0	5,200	3,400
3	16	1,500	0.0	2.7	42.0	680	920
4	11	2,100	4.0	2.8	32.0	830	910
5	33	26,000	14.0	2.4	71.0	16,000	16,000
6	15	1,400	0.0	0.2	330.0	810	930
7	65	3,000	53.0	1.2	280.0	3,700	1,500
8	1	950	0.6	0.5	1.4	1,010	330
9	57	2,600	36.0	3.6	2,700.0	5,300	2,200
10	21	1,100	6.6	0.8	250.0	2,500	860
11	31	1,200	7.5	4.2	390.0	2,400	1,900
12	18	1,200	0.5	0.1	1.2	930	1,100
13	46	850	5.0	1.9	5.6	850	970
14	36	1,300	2.9	---	17.0	1,300	1,100

Table 20. MILL EFFLUENT NO. 3

Sample No.	Ra-226 pc/l	U µg/l	Pb-210 pc/l	Po-210 pc/l	Th (alpha) pc/l	Gross alpha pc/l	Gross beta pc/l
1	15.0	2,500	22.0	21.0	107,000	72,000	90,000
2	4.4	1,900	36.0	6.1	111,000	100,000	65,000
3	7.3	1,900	6.8	5.5	87,000	109,000	37,000
4	13.0	2,300	1.5	6.2	116,000	170,000	46,000
5	28.0	2,900	45.0	11.0	83,000	95,000	51,000
6	26.0	1,400	62.0	30.0	92,000	104,000	36,000
7	40.0	2,000	260.0	170.0	85,000	99,000	52,000
8	20.0	2,900	39.0	21.0	135,000	130,000	71,000
9	25.0	1,600	120.0	110.0	76,000	106,000	33,000

Table 21. MILL EFFLUENT NO. 4

Sample No.	Ra-226 pc/l	U µg/l	Pb-210 pc/l	Po-210 pc/l	Th (alpha) pc/l	Gross alpha pc/l	Gross beta pc/l
1	31.0	1,900	9.0	14.0	29.0	610	740
2	16.0	1,200	4.0	2.2	30.0	400	470
3	0.8	1,100	1.0	0.7	0.0	370	210
4	1.5	1,400	0.9	0.7	0.4	1,400	210
5	16.0	1,700	13.0	1.8	1,500.0	3,325	1,880
6	2.4	1,000	13.0	18.0	17,000.0	21,000	6,900
7	3.2	2,000	140.0	30.0	28,000.0	43,000	15,000
8	9.5	1,800	200.0	110.0	9,700.0	13,000	4,600
9	22.0	1,100	1.6	1.4	32.0	1,100	670
10	31.0	900	63.0	22.0	44.0	980	610
11	0.5	640	0.7	0.3	1.8	600	360
12	0.9	1,700	19.0	56.0	3,800.0	6,100	1,600

Table 22. MILL EFFLUENT NO. 5

Sample No.	Ra-226 pc/l	U µg/l	Pb-210 pc/l	Po-210 pc/l	Th (alpha) pc/l	Gross alpha pc/l	Gross beta pc/l
1	28	7,800	4.3	3.4	69.0	6,800	3,000
2	38	1,800	20.0	19.0	64.0	1,500	1,300
3	65	470	1.5	2.0	60.0	710	430
4	25	980	1.9	2.0	8.6	1,100	750
5	25	980	6.8	4.8	3.0	1,100	1,100
6	28	330	0.7	1.5	31.0	390	540

Table 23. MILL EFFLUENT SUSPENDED SOLIDS

Sample No.	Mill No.	Suspended solids grams per liter	Ra-226 pc/g	Ra-226 pc/l
1	A	0.0195	3,500	190.0
2	A	0.1190	852	101.0
3	A	0.0120	1,809	22.0
4	A	0.1042	1,400	140.0
5	B	0.0427	25	1.1
6	B	0.0318	260	8.4
7	B	0.0367	230	8.4
8	B	0.0376	2,045	77.0
9	B	0.0240	238	5.7
10	C	0.0534	800	16.0
11	C	0.5110	72	37.0
12	C	0.1688	837	141.0

SECTION X

WASTE TREATMENT AND DISPOSAL

Waste retention and treatment practices for control of pollution have improved greatly since 1960. It was common practice in the 1950's to release the spent ore solids and liquid waste directly to unrestricted areas. Most of the mills operating at that time were constructed near streams and the waste discharged directly into the streams.

As a result of the Animas River survey which provided knowledge of the fate of radionuclides in the water, remedial measures were established to confine the contaminants.[29] The measures included removal by sedimentation of the greater part of the spent ore solids, removal of at least 70 percent of the dissolved radium from the effluents, and removal of the toxic chemicals that were destroying the aquatic life for many miles downstream. Initiated at the Durango mill on the Animas River, the measures were in operation by late October, 1959. An evaluation survey, conducted in November, 1959, verified the immediate benefits of the abatement procedures.

Radioisotope concentrations in water vary in the degree of hazard and have been given a maximum permissible concentration in water (MPC_w) by various committees.[30, 31] The standards will be discussed in a later section of the report. Table 24 lists each of the members of the uranium-radium family in order of increasing maximum permissible concentration in water.

Radium-226 has the lowest maximum permissible concentration, 3.3 picocuries per liter, indicating that it is considered to be the most hazardous of all the waste products. Picocuries of radium-226 may be expressed as picograms by applying a 1.02 multiplication factor. As shown in Table 24, radium is a bone-seeking alpha emitter with a half-life of 1,620 years.

Table 24. URANIUM-RADIUM FAMILY, MPC_w VALUES[a]

Nuclide	MPC_w pc/liter	Critical organ	Half-life	Emission
Ra^{226}	3.3	Bone	1,620 yr	Alpha
Pb^{210}	33.0	Kidney	22 yr	Beta
Po^{210}	233.0	Spleen	140 days	Alpha
Th^{230}	667.0	Bone	8×10^4 yr	Alpha
Th^{234}	6,667.0	GI tract	24.1 days	Beta
U^{234}	10,000.0	GI tract	2.5×10^5 yr	Alpha
U^{238}	13,300.0	GI tract	4.5×10^9 yr	Alpha
Bi^{210}	13,300.0	GI tract	5 days	Beta
Pa^{234}	b	---	1.1 min	Beta
Po^{218}	b	---	3.05 min	Alpha
Po^{214}	b	---	1.6×10^{-4} sec	Alpha
Bi^{214}	b	---	19.7 min	Beta
Pb^{214}	b	---	26.8 min	Beta
Rn^{222}	(gas)	Lung	3.8 min	Alpha

[a] MPC_w value is the Maximum Permissible Concentration in water for an average member of the general population (1/30th HB69 value for continuous occupational exposure).

[b] No value given for these short-lived materials.

The nuclide does not precipitate from solution as readily as the other isotopes and is rapidly leached from suspended waste material, thereby contributing to the dissolved activity of water.

The MPC_w for uranium of 13,300 picocuries per liter is equivalent to 40 milligrams of uranium per liter. Chemical toxicity of uranium, rather than radioactive hazard, is the determining factor for the high maximum permissible concentrations permitted. Concentrations in the mill waste are routinely less than this value since extractive methods employed in the milling process are extremely efficient.

Lead-210 and polonium-210 have sufficiently long half-lives and a low MPC_w to warrant consideration as potential pollutants. The isotopes are related in that polonium-210 concentrations in waste are dependent on the lead-210 present. The polonium-210 will reach equilibrium again with the lead-210 in a little over a year even if the two isotopes have been separated in the process. When lead-210 is absent, polonium-210 present will decay almost completely in a year. Little is known of the fate of the two nuclides through the milling process, but Tables 18 through 22 show that concentrations for lead-210 range from less than 1.0 to 1,250 picocuries per liter of effluent. Polonium-210 concentrations have been found to be somewhat less, possibly due to difference in precipitation characteristics and lack of equilibrium. Lead-210 is the most hazardous of the two radionuclides with a MPC_w of 33 picocuries per liter.

Of the two thorium isotopes, thorium-230 is of the greatest concern in mill wastes since it is a bone-seeking alpha emitter with an extremely long half-life. The MPC_w for thorium-230 is 667 picocuries per liter. Thorium compounds are insoluble at neutral or higher pH levels and are discharged primarily in the solid waste material in the alkaline leach milling process. The acid leach process will dissolve a considerable amount of thorium into the liquid waste. Tables 18 through 22 indicate a range in the concentration of alpha-emitting thorium isotopes from less than 1.0 to 135,000 picocuries per liter in a variety of mill effluents from acid leach processes.

Bismuth-210 has been assigned a MPC_w of 13,300 picocuries per liter, but along with other nuclides shown in Table 24, has not been considered significant as a pollutant due to the short half-life.

Due to variances in waste treatment circuits, the radionuclide content of the effluent may be highly inconstant as seen in Tables 18 through 22. No data is available for a continued period of time for an alkaline leach mill effluent.

Waste treatment systems have been designed with the major objective to remove extremely hazardous radium-226 from the effluent. Various types of waste treatment and disposal practices will be discussed.

CONTAINMENT OF WASTES

All solid and liquid waste from the mill is completely contained within a tailings pond providing sufficient land area is available. The size of the ponds vary from a few acres to over 100 acres, and number from one to ten or more at individual mills. The ponds are constructed by building earthen dikes around the disposal area. Soil of a bentonitic nature is preferred for a tailings pond area to reduce seepage of waste into the ground. As construction progresses, the earth is compacted to make the dikes more impervious to seepage. The dikes are usually ten feet across the top and ten to twelve feet high. Some mills separate the sands and the slimes by means of cyclone separators. The coarse sand tailings are used to increase the height of the dikes as the ponds are filled, while the slimes are discharged to the inner area of the pond to serve as a sealant against seepage. With the waste contained in the tailings pond area, all liquid loss is through evaporation or seepage into the ground. Seepage loss has been estimated to be no greater than seven percent by one mill in a pond with a large buildup of slimes on bottom.

The evaporation-percolation pond has been used for tailings disposal following chemical treatment of the wastes.[32] This type of pond is used where land area is not available for large ponds and discharge into nearby streams is not desirable. Seven of these ponds have been built by the

Union Carbide Corporation at Uravan, Colorado. The soil is porous and estimates have been made that evaporation has accounted for 15 percent and percolation 85 percent of the losses. A small amount of seepage, less than 50 gallons per minute, has been observed to reach the river. A decrease in percolation rate has been noted since construction, indicating an expected gradual drop in the disposal capacity due to sealing from the deposition of tailings.

The solid waste accumulates at a rate of approximately 1,960 pounds per ton of ore processed, and by the close of 1969 the total accumulation of tailings in the United States amounted to 83 million tons.[33] Nine million tons per year of tailings were being produced by mills in 1971. The tailings are composed of coarse sands and fine solids or slimes. The fine solids contain more radium-226 per unit weight and are the most easily distributed by wind erosion.

The prevention of direct discharge of waste solids to surrounding waterways has proved to be the most significant pollution control measure instituted by the uranium milling industry. The solids previously served as a long-term source of radium-226 in the stream due to leaching.

The environmental effects of tailings piles have been of concern since 1957. Many studies and meetings have been conducted through the years to identify the hazards, establish rules for controlling tailings disposition, and determine responsibility for stabilization and control of the tailings.[33]

Problems with respect to the contained tailings both during use and after abandonment still exist. These include (1) wind erosion of solids to unrestricted areas, (2) slides into river of piles located near river banks, (3) rising water level during flood conditions to the base of the pile causing leaching of radium from the material, and (4) percolation of water through piles into ground water.

Environmental surveys have been made of several uranium mill tailings piles to evaluate potential radiation hazards.[34, 35, 36] Samples of airborne particles, air, well water, stream water, and tailings material were analyzed for radionuclide content. Geiger survey meters were used to measure gamma radiation from the tailings material and the surrounding area. The conclusions from the studies were that (1) the radiation levels on the tailings were of such levels as to preclude the release of the tailings area for public use, (2) wind erosion had spread tailings material to distances of 1,000 feet from the tailings area to the extent that radiation levels exceeded recommended standards, (3) radium-226 and thorium-230 concentrations in air exceeded recommended concentrations downwind from the pile if left uncovered and unstabilized, (4) radon-222 gas in the area was not a hazard unless enclosed structures were to be built on the material, and (5) well water and stream samples did not show pollution from the tailings area. Recommendations were to stabilize the tailings against wind erosion to eliminate the potential long-term hazard to inhabitants of the area. Also the area should not be released for public use until the tailings were covered with uncontaminated soil to a depth that would lower radiation levels to acceptable limits.

Remedial measures have been undertaken by members of the industry. Tailings piles near river banks are being retrieved and rocks placed along the sides of the pile to prevent water erosion.[32] Other companies have attempted to establish vegetation directly on the tailings material to prevent wind erosion. The procedure has not been highly successful due to the low pH of the material, lack of moisture, and blowing tailings sand which severs the blades of grass soon after sprouting. Most tailings piles are located in arid regions and would require watering to sustain vegetation. A study of plants requiring a minimum of fertilization and ground preparation and possessing the best growth characteristics for long-term maintenance of tailings piles, has been conducted at the Colorado State University.[37] Results have not been encouraging because of the long-term maintenance measures necessary to insure vegetation cover.

An attempt to establish vegetation on the dikes to prevent wind erosion is being made by companies. One company located in a timbered area has planted trees on top of the dikes to prevent wind erosion of the tailings material. Another company sprays tailings solution over the tailings area to stabilize the fine material and prevent dusting. This method would be useful for large tailings ponds with exposed disposal areas not covered by solution and subject to wind erosion.

Stabilization of the Monticello, Utah, tailings pile was accomplished by covering with two feet of soil and planting vegetation at a cost of $5,000 per acre.[38] Another report states that coverage with one foot of soil would cost $1,100 per acre.[39] The costs vary depending on the location of pile in relation to soil utilized for covering, and the labor involved in moving tailings solids for consolidation and contouring. Estimated costs, ranging from $1,360 to $2,910 per acre, have been calculated for several Colorado River Basin tailings piles by an earth-covering method and a chemical-covering method.[39] A chemical and vegetative method of application in which the Bureau of Mines was involved cost $135.50 per acre.[38] Several means for stabilization of relatively fine-sized waste, such as those contained in uranium mill tailings piles, have been investigated by the Bureau of Mines.[38]

The Bureau of Mines assisted the El Paso Natural Gas Company in devising a satisfactory and reasonable cost method of stabilizing the uranium wastes at Tuba City, Arizona. Vegetation procedures were not considered since less than nine inches of precipitation per year is insufficient to maintain continuous coverage. Due to cost considerations, physical stabilization using rock and soil coverings was considered impractical. From 20 chemicals tested for stabilization ability, an elastomeric polymer and a lignosulfonate were selected for use on the Tuba City tailings. The polymer, due to its deeper penetration properties (approximately two inches), was applied to the tailings pond dike as this area was subject to the greatest danger of wind and water erosion. The lignosulfonate, having a penetration depth of only three-fourths inch but resulting in a surface with a hard crust, was sprayed on the surface of the tailings material contained within the dike. Both laboratory and field tests revealed no signs of wind erosion following application of the chemicals. Estimated costs of the treatment were $335 per acre.

CHEMICAL TREATMENT AND DILUTION

Waste discharge for acid and alkaline leach processes average 1,000 and 250 gallons per ton of ore respectively. Recycling may reduce the total volume; however, ultimate disposal of the waste liquor must occur by evaporation, seepage into the ground, or discharge into rivers or streams.

From 1.3 to 3.9 acres of pond surface are required to evaporate one ton of water per hour depending on rainfall conditions in the Western United States.[4] Assuming no loss through seepage and a factor of 2.6 acres per ton of water evaporated, a 1,000 ton per day mill using 500 gallons of water per ton of ore would require a 220 acre pond. At 1,000 and 250 gallons of water per ton, a 440 and 110 acre pond would be required. Ponds that allow significant quantities of waste liquor to seep into the ground require proportionately smaller pond areas.

Mills located in areas where sufficient land is not available to contain the total effluent treat the effluent to reduce the level of radioactivity, and release controlled amounts into nearby waterways. Neutralization of the effluent to a pH of about eight effectively removes 90 percent of the dissolved radium, essentially all of the thorium, and some inorganic contaminants. The radium-226 content of untreated effluent is in the range of 500 to 1,000 picocuries per liter; following neutralization, the range is from 50 to 100 picocuries per liter.

Initially, barium sulfate (barite) was used to remove radium-226 from the effluent, but has been replaced by barium chloride due to its greater efficiency.[40] The radium is co-precipitated with barium sulfate upon addition of barium chloride at a rate of 0.05 to 0.3 gram per liter to the effluent. Under optimum conditions, 99 percent of the radium is removed. The precipitate is allowed to settle before the treated effluent is discharged to a river or stream. Dilution effects further reduce the concentration of contaminants.

The Union Carbide mill at Uravan, Colorado, has rearranged the barium chloride decontamination circuit to combine ditch drainage with the plant effluent and to continuously monitor the pH of the effluent. The circuit modifications have reduced the radium-226 concentration in the effluent significantly. One industrial company producing uranium and thorium employs a sodium hydroxide neutralization process to a pH greater than eight and two barium sulfate precipitations to reduce the radium concentrations by 99.9 percent.[41] Other laboratory studies have shown that radium-226 can be removed efficiently from simulated lime-neutralized acid waste by adsorption on a number of inorganic ion exchange materials.[42] Anomalies in the radium concentration present in discharges have been noted.[32] Variations in radium-226 content of effluents may be seen in Tables 18 through 22 and in the suspended solid content in Table 23. Apparently, the efficiency of decontamination is dependent upon the initial effluent concentration, variations in the treatment process, and retention time in the settling ponds.

Mine waters, containing radium-226 concentrations as high as 192 picocuries per liter, are released to unrestricted areas at rates up to 3,000 gallons per minute at some sites; settling ponds are utilized to collect suspended solids prior to mine water discharge.

UNDERGROUND DISPOSAL

Underground disposal of wastes has been used by the petroleum-producing industries since 1920 for oil field brine. More industries have begun using this means of disposal through the years so that by 1962 approximately 30 underground disposal systems were in operation. By 1969 almost 200 wells were in use.

One currently operating uranium mill, the Anaconda Company, is disposing of a portion of its effluent by deep well injection.[43] Drilling began in 1959 and required 110 days for completion and testing. Cores were taken to a depth of 2,511 feet and tested to determine the best disposal zone. The well is located near Grants, New Mexico, and passes through the San Andres and Glorieta fresh water aquifer at a depth of about 360 to 500 feet. Impermeable formations lie between the fresh water aquifer and the disposal zone. The well was completed in 563 feet of sandstones in the lower San Ysidro and Meseta Blanca members of the Yeso formation. The disposal zone lies in the sandstone at a depth of 940 to 1,420 feet.

The well was cased to seal off all of the fresh-water aquifers and to support the walls of the hole throughout the disposal zone. Surface casing was installed to a depth of ten feet below fresh water, and injection casing to a depth of 1,830 feet in an 11-inch hole. Perforating and fracturing operations were performed in the disposal zone.

The effluent is pumped from a wooden decanter box in the tailings pond in order to minimize suspended solids. Diatomaceous earth is added to the effluent to aid filtration, sodium polyphosphate to retard precipitation of calcium sulfate, and copper sulfate to control the growth of microorganisms. The effluent is filtered to remove suspended solids, thus preventing plugging of the disposal zone. Wastes were injected at an average rate of 80 gallons per minute but rates as high as 600 gallons per minute were experienced.

Since laboratory tests proved that the disposal zone material would neutralize 390 gallons of waste per cubic foot without a loss of permeability, the effluent is not neutralized prior to injection. As the disposal zone material neutralizes the effluent, all thorium-230 and 90 percent of the radium is precipitated.

From 1960 through 1968, 600 million gallons of effluent were injected into the well. Table 25 shows the gallons of effluent, picocuries of radium-226 per liter of effluent, and total curies per year of radium-226 injected from 1964 through 1969.

The capacity of the disposal zone was calculated to be two to four billion gallons. Should the current injection rate continue, the disposal well has a calculated life to the year 1997.

Table 25. WASTE DISPOSAL BY DEEP WELL INJECTION

Year	Gallons of waste	Radium-226 pc/l of waste	Radium-226 curies per year
1964	57,814,250	181	.0476
1965	24,274,342	128	.0141
1966	29,115,770	79	.0104
1967	24,079,220	109	.0119
1968	50,345,550	68	.0156
1969	63,459,275	205	.0592
Total	249,088,407		.1588

RECYCLING OF DISCHARGES

Recycling of the effluent discharge is practiced to reduce the total volume of wastes for disposal and to conserve water in areas of low water supply. Mill solutions are recycled within the plant with only a small portion transferred to the waste disposal area to prevent buildup of interfering substances.

In other instances the total waste liquid is pumped to the tailings pond and allowed to settle. Clear effluent is pumped back to the mills to be reused in the circuits or to slurry leached tailings solids prior to pumping to the disposal area. One mill discharges waste at a rate of 550 gallons per minute while returning clarified waste to the mill for reuse at a rate of 300 gallons per minute. The limiting factor on reuse of waste liquor is the dissolved solid content which may interfere with the milling process.

OFF SITE DISPOSAL

The uranium milling process discards greater than 97 percent of the ore processed as waste mill tailings. Due to the great hazard to health posed by radium-226, a careful plan providing for the safe disposal of tailings is a necessity. Several solutions have been proposed for the disposal problem. Initially, the wastes were to be used for such purposes as land fill and highway subgrade fill material.[39, 44]

In 1966 radon film badges were placed inside newly completed buildings in Grand Junction, Colorado, in an area where uranium mill tailings had been used as fill under and around the building. The badges did not provide the sensitivity for quantitative results, but did show evidence of elevated radon gas concentrations in the buildings. The State of Colorado ordered a halt in the use of tailings material for building projects without prior approval by the Department of Health.[33]

The radon-222 gas being formed from the decay of radium-226 in the tailings can diffuse through flooring material and concrete, and accumulate inside buildings to excessive levels due to inadequate air circulation. The short-lived daughters of radon-222 (polonium-218, lead-214, bismuth-214, and polonium-214) are deposited as solids.

An intensive survey of all buildings built on tailings pile material was begun to determine the level of activity.[33] An estimated 150,000 to 200,000 tons of tailings had been distributed through Grand Junction between 1953 and 1966. The tailings had been used primarily under concrete slabs and around foundations. Other uses had been under streets, driveways, swimming pools, water pipes, and sewer mains. Radon levels exceeded the screening value in 42 percent of the residences and 62 percent of the businesses.

The study and evaluation of the problem has continued with involvement of personnel from the Atomic Energy Commission, U.S. Public Health Service, Environmental Protection Agency, and the Colorado Department of Health. On July 27, 1970, the Surgeon General of the U.S. Public Health Service made recommendations of action for radiation levels in dwellings constructed on or with uranium mill tailings. The recommendations were as follows:

External Gamma Radiation	Recommendation
Greater than 0.1 mR/hr.	Remedial action indicated
From 0.05 to 0.1 mR/hr.	Remedial action may be suggested
Less than 0.05 mR/hr.	No action indicated

Indoor Radon Daughter Products	Recommendation
Greater than 0.05 WL	Remedial action indicated
From 0.01 to 0.05 WL	Remedial action may be suggested
Less than 0.01 WL	No action indicated

Of 534 Grand Junction locations sampled, 65 exceeded 0.05 working level (WL) and 30 were higher than 0.1 mR/hr. The Surgeon General stated that protracted exposure at the upper level rate of 0.1 mR/hr. and 0.05 WL could double the risk of leukemia and lung cancer.

Remedial action studies are being conducted relating to use of tailings in construction projects.[33] The average cost of removing the tailings from under and around the structures has been estimated to be $3,220 per residence. Additionally, studies are under way to find methods of controlling the diffusion of radon through structures.

Radon surveys are being undertaken in other locations where uranium tailings have been utilized for construction purposes. Elevated levels of radon-222 were found in some Uravan, Colorado homes and two families were moved to other housing. Mobile gamma scanning operations are being conducted around uranium milling areas by personnel from the Western Environmental Research Laboratory in Las Vegas, Nevada, in an effort to locate suspected areas. The problem of radon-222 gas emanating from tailings material demonstrates the importance of utilizing effective disposal and containment of tailings material.

One solution to the problem of long-term disposal would involve placement of the tailings back into the abandoned mines. Open pit mines could be filled and covered with topsoil. Some land owners in Texas have specified in the lease that the pit be filled and topsoil replaced to restore land to its original condition. This method may prove to be the most effective and economical for the long term.

SECTION XI

EFFECTS OF WASTES ON THE ENVIRONMENT

Radioactivity measurements of four types of samples--water, sediment, biota, and crop--are useful in demonstrating the degree of pollution resulting from uranium operations. The few mills discharging wastes to rivers and streams at the present time treat the waste prior to discharge; however, mine water containing pollutants is discharged in several mining locations without chemical treatment. Surface water monitoring will immediately indicate any pollution problems resulting from the two types of discharge. The majority of the mills contain all their wastes in tailings ponds with disposal by evaporation or seepage. Seepage from ponds near rivers and streams can be detected by surface water monitoring; however, many tailings ponds are far removed from flowing rivers and streams. To demonstrate pollution in these areas, activity of ground water samples must be measured.

Sediment samples reflect the accumulation of radioactivity over a period of time depending on the flow history of the stream. During periods of low flow, the concentration of activity increases in a polluted location due to the accumulation of mill tailings. During periods of high flow, the contaminated sediment is diluted by upstream sediment and distributed downstream.[45] Radium-226 is leached from the sediment by the scouring action of the water. Sediment samples from reservoirs reflect the extent of pollution over a longer period since the sediments are relatively undisturbed by the reservoir waters. Trends in levels of radioactivity in reservoirs are gradual, but useful in demonstrating the long-term effects from upstream pollution.

River biota (algae, bottom animals, and fish) accumulate and concentrate radium-226; hence, serve as cumulative indicators of pollution. Due to their concentration ability, the organisms are a more sensitive indicator of prolonged pollution than water samples collected intermittently. Plants also accumulate and concentrate radium-226, and edible crops that have been irrigated with contaminated water contribute to the total radioactive intake of the population. Dairy cattle fed on alfalfa, a plant that concentrates radium to a high level, may contain the radionuclide in their milk.[46]

To determine the effect of wastes on the environment, background surveys are extremely important. In many instances the mills producing waste are located near mining areas where large quantities of ore are located in the ground water table. Without knowledge of radiation levels prior to milling operations, the degree of pollution is difficult to assess. Radium and uranium levels may be quite high in ground waters from natural leaching of ore bodies.[47] Levels of natural radioactivity in ground waters in excess of 3 picocuries of radium-226 per liter have been found in several areas of the United States.[48, 49]

In the following discussion, data from previous surveys and ongoing monitoring programs for both radioactive and inorganic pollutants resulting from uranium mining and milling operations will be grouped by sample types. In many instances, data for a particular area are extremely limited.

GROUND WATER

The Gas Hills region of Wyoming is an arid, sparsely populated area. Four uranium mills operate in the region with the mill effluent contained in tailings ponds and disposed of by natural evaporation and seepage. The mills monitor water wells in the vicinity of the tailings ponds to detect radioactive and inorganic pollution. Levels of radium-226 have been detected as high as 50 picocuries per liter; however, the majority of levels are less than 10 picocuries per liter. Seepage has been noted near some tailings ponds by the appearance of surface ponds in low areas near the tailings pond. The seepage water is checked for contaminants by the company and pumped to restricted areas if the concentration of radioactivity is above the mill discharge limit of 30 picocuries per liter of radium-226.

The Dawn Mining Company at Ford, Washington, discharges waste into an evaporation pond located near a river. The pond is not sealed against seepage and is often dry. Company analyses of water from a monitor well located down-gradient from the pond have not shown contamination of the ground water.

The Cotter Corporation mill at Canon City, Colorado, retains all effluent in tailings ponds with all loss of liquid resulting from evaporation or seepage. Seepage has been noted and well waters of high mineral content unfit for livestock consumption have been reported by residents of the area. Contamination from molybdenum is suspected and is under investigation by the Colorado State Health Department. The company operates monitoring wells and sampling stations on the Arkansas River.

A monitoring well below the tailings pond area at Uravan, Colorado, has shown slightly higher radium-226 concentrations and a high mineral content compared to a well up-gradient from the ponds. Two abandoned mines on Mesa Creek have a high radium-226 concentration in the drainage water that seeps into the ground and drains into the river. No data on ground water is available for the area. Several large trees have died down-gradient from the tailings area at Rifle, Colorado, indicative of a high mineralization in the ground water.

Mill operations at Monticello, Utah, have been terminated and the tailings pile stabilized against erosion. Ground water samples taken in the area in 1967 showed radium concentrations as high as 17 picocuries per liter possibly due to leaching of radioactivity from the pile.

Three uranium mills are located in the Grants, New Mexico area. The area is semi-desert and the creeks and washes are usually dry except during flash floods. Rio San Jose, the nearest river, is intermittent in flow. Studies indicate that the background concentration of radium-226 in the Grants area ranges from 0.1 to 0.4 picocuries per liter. Two wells near San Mateo Creek, which receives mine water discharges and runs near the milling areas, had levels of radium-226 greater than background in 1962. Mine drainage water contained 345 picocuries per liter of radium-226.

An investigation was made in 1956 to determine whether the ground water in the Grants-Bluewater area was being polluted by uranium waste stored in a tailings pond.[50] From a total waste discharge of 4.3 million gallons per day, seepage losses were calculated to be 0.3 million gallons. The loss required a seepage rate of 0.17 feet per day from the 70 acre pond. The pond had been in use for three years at the time of the survey and mill personnel anticipated that slimes in the waste would eventually seal the pond. The investigation revealed that all inorganic constituents other than nitrates tended to be fairly constant. Nitrates increased the nearer the ground water was sampled to the tailings area with the highest value being 16.9 milligrams per liter of nitrate-N. Findings elsewhere have shown that nitrates travel more rapidly in soils than do other constituents due to the selective nature of the ion exchange capacity of the soil. As the ion exchange capacity of the soils is exhausted, the other constituents, both inorganic and radioactive, may be found at greater distances from the source of pollution.

The company has changed the milling process to eliminate the use of nitrate compounds and has begun disposing of the greater part of the effluent by injection to a deep well. It was believed that this disposal method would reduce contaminants to a minimum in the ground water system. The company operates monitor wells on the property and samples wells in the surrounding area to detect any changes in the water quality. Analyses of monitoring well samples by the company from 1964 through 1969, showed radium-226 concentration greater than two picocuries per liter at only four locations. The highest radium-226 concentration was 5.2 picocuries per liter, demonstrating an upward trend at the location from a concentration of less than two in 1964, to 5.2 in 1969. Nine sampling locations showed a gross alpha concentration somewhat above background levels, but no definite trend over the six-year period was established.

The other two mills in the Grants area operate monitoring wells at depths of 75 to 100 feet near the tailings pond areas. All three mills are located in close proximity, making pollutant source identification difficult. The radium-226 concentration has ranged from 1 to 25 picocuries per liter in one company's well.

Texas Gulf Coastal Plain operations employ open pit mining in which the ground water is pumped from the mine continuously and mill wastes are totally contained in tailings ponds. The uranium mills and the Texas State Health Department have sampled wells in the mining and milling area for radioactivity. Results from well to well have varied widely in radium-226 concentration. In the majority of instances, the concentrations were in excess of ten picocuries per liter with some values considerably higher. Background data is not available to assess pollution from the mining and milling operations. It is difficult to determine whether elevated levels of contaminants are a result of mine and milling activities or natural leaching of ore bodies in the area. The Continental Oil Company is collecting and analyzing surface water and soil samples from a future milling area to establish a background data for determining normal variations.

SURFACE WATER

Since 1950 river water samples have been analyzed from the Colorado River Basin to detect the presence of radium-226 in the environment resulting from uranium milling operations.[51] At that time the radioactive pollution of the water was extensive, but the degree of hazard to the public was not fully realized. Radium concentrations ranged from a background level of 0.2 picocuries per liter to 43 picocuries per liter in mining and milling areas. Most surface waters affected by the uranium mill discharges were located in the Colorado River Basin.

In September, 1955, a preliminary field study was made to evaluate the extent of stream pollution by waste discharges from uranium mills. The effluents from the mills contained soluble radium-226 ranging from 4.5 to 920 picocuries per liter and large quantities of suspended tailings solids high in radium-226. Water samples taken from below the mills had radium concentrations up to 86 picocuries per liter compared to average background levels of 0.3 picocuries per liter. A more detailed survey was made in 1956 that substantiated the earlier findings. Leaching of radium-226 from river muds and mill tailings solids by stream water and uptake of the radionuclide by the stream biota was noted. Further work on the ultimate fate of radium-226 in the stream environment was recommended.

The serious nature of the problem was realized in 1957 when the standards for exposure were revised downward and the U. S. Atomic Energy Commission published regulations for waste discharge by licensees.[52] As a result of a conference held in 1958, a one-year fact-finding survey was performed to assess the interstate pollution of the Animas River by uranium mill waste discharge.[46]

The Animas River survey showed the dissolved radium-226 content of mill waste discharge to range from 44 to 822 picocuries per liter. An estimated 30 to 40 milligrams of undissolved radium-226 was being discharged daily in 15 tons of tailings solids. At locations of 2, 23, 28, and 59 miles below the mill, radium-226 concentrations were 12.6, 7.2, 7.6, and 2.9 picocuries per liter, respectively. The radium-226 concentration in raw water supplies of Aztec and Farmington, New Mexico, averaged 3.6 and 2.8 picocuries per liter. Treated water from the municipalities averaged 3.6 and 2.6 picocuries per liter. At times treated water samples contained more radium-226 than the raw water. The apparent anomaly was explained by the leaching effect of the water on water treatment plant filter sands polluted with radium-226. Concentrations of radium-226 ranging from 25 to 30 picocuries per gram were found in the Aztec, New Mexico, water treatment plant filter sands. The radium-226 was present in the form of suspended solids or as a consequence of adsorption of dissolved radium-226 on the sands. An average radium-226 concentration of 7.6 picocuries per liter in raw water was being used by New Mexico residents during the survey.

The dissolved radium-226 released in the mill effluent discharged to the river could not account for all the dissolved radium-226 at a station two miles below the mill. The bulk of the mill solids discharged settled on the river bottom to serve as a source for continued leaching into the water. Periods of high river flow carried the solids downstream.

As a result of the survey data which implicated solid waste tailings discharged to the river as a major source of dissolved radium, laboratory investigations were performed to define the degree and conditions associated with leaching.[53, 54, 57] Conclusions from these investigations were as follows:

1. No appreciable amount of radium-226 leaching occurred after 30 minutes with periods of time up to six days.

2. The liquid to solid ratio was one of the more important variables. Little increase in leaching occurred at ratios greater than 1,000: 1, but up to this ratio, the leaching rate increased rapidly.

3. The amount of radium-226 leached is dependent on the quantity of radium-bearing solids (or total radium reservoir) after a liquid-solid ratio greater than 1000: 1 is reached.

4. Subsequent leachings of mill tailings were not effective as seen by Table 26. Storing solids under wet conditions for a period of time prior to releaching did not result in increased radium leaching.

5. The fine ore particles contained the greater quantity of radium-226.

6. Barium chloride greatly enhanced the removal of radium from tailings. After maximum leaching with water, a 0.01 molar barium chloride solution leached about 35 percent additional radium-226. Barium was the only common inorganic element tested that caused significant leaching.

7. The sulfate and barium content of the solids influences the amount of leached radium-226 that will remain in the dissolved state. Dissolved radium-226 co-precipitates as a barium-radium sulfate when sulfate and barium ions are present in sufficient quantities.

8. The leaching effect is greater for uranium mill waste solids than for river sediments and for acid leached waste solids than for alkaline leached solids.

A followup survey was made in November, 1959, after treatment facilities were installed in the Durango uranium mill.[55] The treatment procedures reduced toxic chemicals by 76 percent, dissolved radium by 80 percent, and removed 89 percent of the suspended ore solids. Substantial improvement in stream water quality was made by the installation of the treatment procedures.

Table 26. REPETITIVE LEACHING OF MILL TAILINGS

Time	Percent radium-226 leached
30-min. initial leaching	1.61
1st additional hour	0.17
2nd additional hour	0.05
3rd additional hour	0.07
60-min. initial leaching	1.55
1st additional hour	0.14
2nd additional hour	0.09
3rd additional hour	0.24
120-min. initial leaching	1.24
1st additional hour	0.23
2nd additional hour	0.13
3rd additional hour	0.05

As a result of the previous field and laboratory studies, the Colorado River Basin Water Quality Control Project was established in 1960. Figure 11 illustrates the location of the Radium Monitoring Network sampling stations, while Table 27 lists the mean annual concentrations of radium-226 in water at these stations from 1961 through 1970.

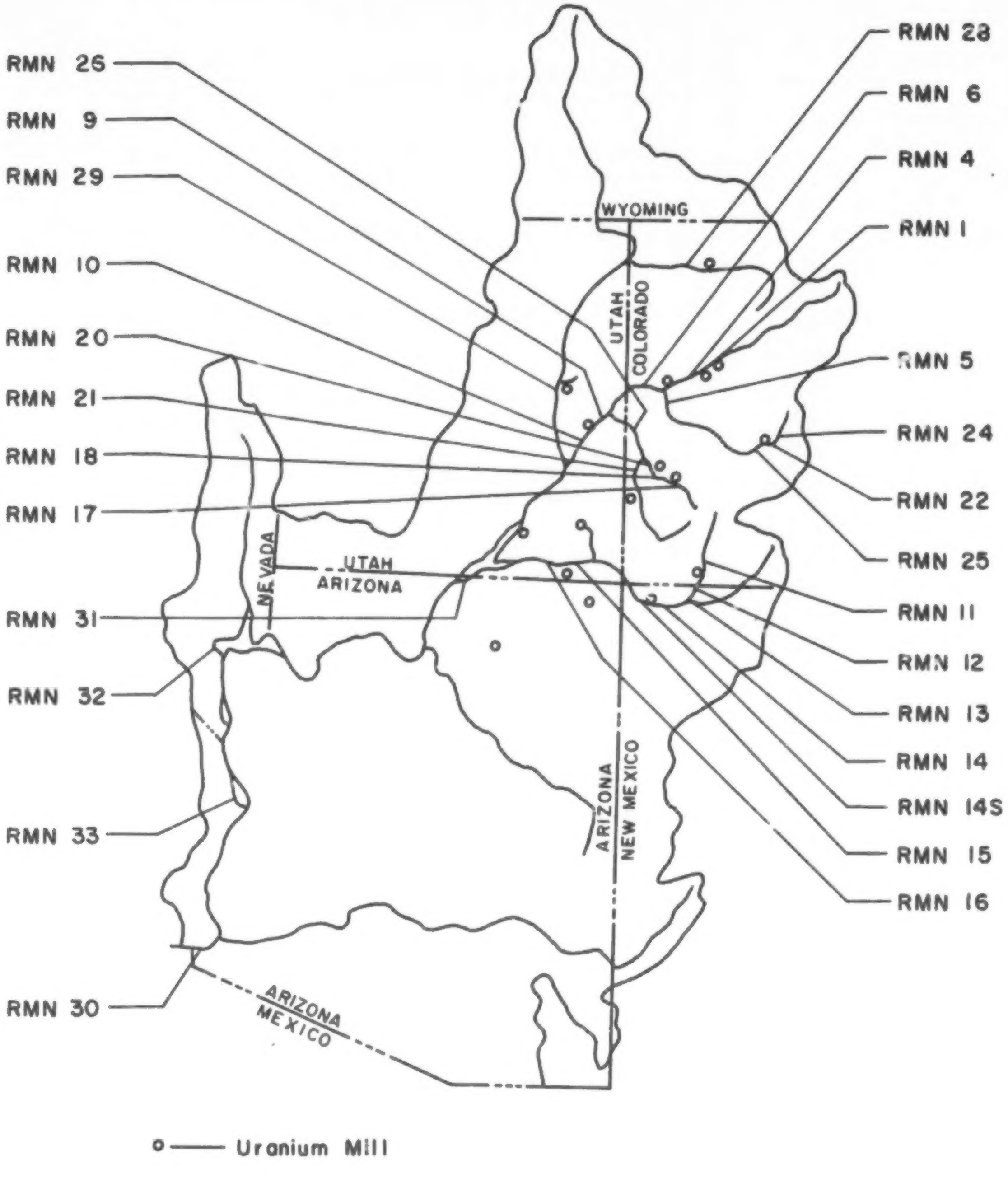
The Durango mill operated from 1959 to 1963; following closure of the mill in March, 1963, the radium-226 concentration at Station 12 below the mill decreased to near background levels by 1965. Effects from the Uravan milling operations are shown by the elevated and fluctuating radium-226 concentrations at Station 20 located immediately below the mill. Concentrations at Station 20 ranged from 3 to 30 times higher than above the mill. Individual sample values varied widely from 26.0 to 0.5 picocuries per liter for a 3.94 average during one 6-month time interval in 1968-69.

Radium-226 polluted water from the San Miguel River and mine drainage from Mesa Creek enters the Dolores River upstream from Station 26, thus accounting for the elevated values measured at that station.

Concentrations of radium-226 at Station 9 below the confluence of the Dolores and Colorado Rivers are consistently higher than at Station 6 upstream. Also, values at Station 10 downstream from the Moab Mill are higher than at Station 9 upstream from the mill. The greatest concentration difference between Stations 9 and 10 was 0.65 picocuries per liter.

Radium-226 concentration in Lake Mead and Lake Havasu, Stations 32 and 33, are consistently higher than at Station 31 upstream and Station 30 downstream. Leaching of naturally-occurring uranium ore bodies in the lake areas may contribute to the elevated values found.

Other uranium mining areas have not been studied to the extent of the Colorado River Basin. All the uranium mills conduct monitoring programs to satisfy the requirements of their license. State and government agencies have performed limited investigations in the Shirley Basin of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RMN 13
RMN 14
RMN 14S
RMN 15
RMN 16
WYOMING
UTAH
COLORADO
NEVADA
UTAH
ARIZONA
ARIZONA
NEW MEXICO
ARIZONA
MEXICO
o—— Uranium Mill

Figure 11. COLORADO RIVER BASIN RADIUM MONITORING NETWORK

Table 27. MEAN ANNUAL CONCENTRATIONS OF RADIUM-226
IN WATER AT MONITORING NETWORK STATIONS, 1961-1970
(values in picograms per liter [1.02 picograms = 1 picocurie])

RMN Station		Years									
No.	Location	1961	1962	1963	1964	1965	1966	1967	1968	1969	1970
1	Colorado River at Silt, Colorado	0.18	0.15	0.23	0.25	0.16	0.20	0.20	0.19	0.17	0.17
4	Colorado River above DeBeque, Colorado	0.20	0.16	0.23	0.22	0.17	0.18	0.19	0.18	0.14	0.11
5	Gunnison River above Grand Junction, Colorado	--	0.07	0.11	0.12	0.07	0.09	0.08	0.08	0.07	0.05
6	Colorado River near Fruita, Colorado	--	0.17	0.21	0.18	0.17	0.16	0.17	0.14	0.11	0.10
9	Colorado River above Moab, Utah	0.34	0.24	0.29	0.27	0.19	0.26	0.28	0.24	0.17	0.15
10	Colorado River below Moab, Utah	0.41	0.33	0.54	0.32	0.27	0.39	0.93	0.45	0.20	0.18
11	Animas River at Durango, Colorado	0.05	0.05	0.06	0.05	0.04	0.05	0.05	0.05	0.05	0.08
12	Animas River at Colorado-New Mexico State Line	0.37	0.37	0.23	0.13	0.09	0.09	0.09	0.07	0.05	0.05
13	San Juan River above Farmington, New Mexico	--	0.08	0.07	0.04	--	--	--	--	--	--
14	San Juan River at Fruitland, New Mexico	--	0.12	0.12	0.07	0.06	0.05	0.06	0.06	0.06	--
14S	San Juan River below Shiprock, New Mexico	--	0.11	0.12	0.09	0.06	0.05	--	--	0.06	0.06
15	San Juan River above Mexican Hat, Utah	0.18	0.15	0.13	0.14	0.07	0.08	0.08	0.09	0.08	0.05
16	San Juan River below Mexican Hat, Utah	0.34	0.24	0.16	0.09	0.07	0.07	0.08	0.07	0.08	0.04
17	San Miguel River above Naturita, Colorado	0.02	0.04	0.04	0.05	0.05	0.05	0.04	0.04	0.04	0.04
18	San Miguel River above Uravan, Colorado	0.14	0.21	0.33	0.14	0.16	0.13	0.12	0.11	0.07	0.09
20	San Miguel River below Uravan, Colorado	0.98	0.84	1.34	0.86	0.46	1.92	1.57	3.37	1.54	0.42
21	Dolores River at Bedrock, Colorado	0.85	0.39	0.45	0.61	0.28	0.33	0.50	0.30	0.28	0.36
22	Tomichi Creek above Gunnison, Colorado	0.02	--	--	--	--	--	--	--	--	--
24	Gunnison River above Gunnison, Colorado	0.03	0.05	0.05	0.05	0.04	--	--	--	--	--
25	Gunnison River below confluence of Tomichi Creek, Gunnison, Colorado	0.04	0.04	0.05	0.04	0.03	--	--	--	--	--
26	Dolores River at Gateway, Colorado	0.42	1.38	1.03	1.13	0.67	1.20	1.22	1.78	1.71	0.59
28	Yampa River below Maybell, Colorado	0.11	0.09	0.09	0.09	0.06	--	--	--	--	--
29	Green River below Green River City, Utah	--	0.06	0.10	0.09	--	--	--	--	--	--
30	Colorado River at United States-Mexican Border	--	0.15	0.17	0.18	0.16	0.16	0.17	0.14	0.13	0.11
31	Colorado River at Page, Arizona	--	--	0.25	0.25	0.17	0.15	0.14	0.16	0.15	0.13
32	Colorado River at (Lake Mead) Boulder City, Nevada	--	--	0.33	0.31	0.41	0.41	0.32	0.22	0.19	0.16
33	Colorado River at Metropolitan Water District Intake, Lake Havasu, California-Arizona	--	--	0.35	0.36	0.36	0.39	0.34	0.26	--	--

Wyoming and the Texas Gulf Coastal Plains. Surface water drainage from Shirley Basin, Wyoming, drains to Spring Creek, Little Medicine Bow River, Seminoe Reservoir, and the North Platte River system. An estimated 1,500 to 3,000 gallons per minute of water from open pit mines and dewatering wells is pumped into Spring Creek. Radium-226 concentrations of 35 picocuries per liter have been detected below the confluence of Spring Creek and the Little Medicine Bow River. Uranium was detected in concentrations of 1,000 micrograms per liter at the same location. Radioactivity in Seminoe Reservoir has remained at background levels. Elevated selenium concentrations have also been found in water samples from the area.

In the Texas Gulf Coastal Plains area mine water was discharged to a creek draining to a river and a lake. Radium-226 and uranium analyses of creek water indicated elevated levels below the discharge point. The chloride content was also above normal. To remedy the problem, the company began discharging the mine water to an abandoned mine.

RIVER SEDIMENT

River sediment is polluted with radioactivity from the uranium industry by the direct discharge of solid waste tailings, by smaller amounts of suspended fines in the discharged effluent, and by chemical precipitation of dissolved radioactive substances by sulfates in the water. River sediments serve as cumulative indicators of long-term contamination of the environment. Removal of radioactive sediments from a specific stream location is slow; in one instance, four years was estimated for concentrations to return to normal background levels following termination of solids discharge to the river.[45]

As a result of the Animas River survey, background levels of normal river sediments were established in the range of two picocuries of radium-226 per gram. Levels of radium-226 in the sediment directly below the point of solids discharge to the river measured 115 picocuries per gram.[46] The levels of activity in the sediment are higher during periods of low flow due to the lack of scouring action of the water. Radioactivity is leached from sediments while the sediment is being transported downstream to become eventually deposited in lakes or reservoirs. Due to the high radium-226 content of the waste solids and the leaching effect of the river water, a significant portion of the dissolved radium has as its source the deposited solids on the river bed. Direct discharge of tailings materials by mills to waterways was banned in 1959.

Fine suspended solids in the mill waste effluent represents the current major source of radium-226 in river sediments. Table 23 lists suspended solids content of mill effluents. Radium-226 values as great as 3,500 picocuries per gram and 190 picocuries per liter of effluent may be seen. Laboratory studies have shown that radium-226 may be leached from solids in amounts as high as 50 percent. Using the above values, 50 percent leaching would produce 95 picocuries per liter dissolved radium-226, an amount considerably greater than is normally present in an effluent following treatment. Removal of suspended solids is necessary to minimize the radium-226 concentration in water.

RESERVOIR SEDIMENT

The ultimate destination of contaminated waste solids is in the bottom sediments of lakes and reservoirs. In an effort to obtain background concentrations of radium-226 in Lake Mead, two core samples of sediment deposited over the years 1935-1949 were analyzed.[59] Radium-226 concentrations ranging from 1.5 to 2.0 picograms per gram were obtained from the core samples according to the year of deposition. After development of the uranium industry, concentrations in surface sediments ranged up to 5.9 picograms per gram. An average of 2.9 picograms per gram was calculated for all sediment samples analyzed for the period from 1960 to 1964.[45] Lake Mohave and Lake Havasu sediments averaged 1.3 and 2.2 picograms per gram, respectively, for the same period. Lake Mead, located upstream from the other reservoirs, has been the primary collecting area for contaminated sediments.

Lake Powell, in southern Utah, replaced Lake Mead as the recipient of sediments from the Colorado River in 1964. Lake Powell sediments were analyzed in 1965, and found to be near background levels. Water samples ranging from 0.18 to 0.26 picograms of radium-226 per liter showed lower concentrations of radioactivity than Lake Mead. At the time of sampling, the lake had not had time to accumulate contaminated sediments. Unfortunately, more recent data are not available.

Only limited sampling has been conducted in other reservoirs located downstream from uranium mining and milling operations. The reservoirs include: Boysen Reservoir, downstream from the Gas Hills operations; Seminoe Reservoir, downstream from the Shirley Basin operations; Angostura Reservoir, downstream from the Edgemont, South Dakota operations; and Lake Corpus Christi, downstream from the Texas Gulf Coastal Plains operations.

AQUATIC BIOTA

Detailed studies of the effects of waste discharge from uranium mills on the aquatic biota of the Animas, Dolores, and San Miguel Rivers were conducted during 1958-63.[46, 55, 56, 60] Prior to the installation of waste treatment processes, essentially all of the benthic organisms were eliminated immediately below the uranium mills, and greatly reduced 35 to 40 miles downstream. Reductions in fish populations were also noted. Release of the raffinate solution from the solvent extraction process in the waste discharge was found to be primarily responsible for the toxicities noted. Installation of treatment facilities resulted in a significant improvement in benthic populations.

Selected benthic invertebrates and attached algae serve as useful cumulative indicators of radioactivity, since both concentrate radium-226 from the surrounding aqueous environment to approximately the same degree.[56] Concentration factors may be calculated from the following formula:

$$C = \frac{\text{radium-226 per gram of live (wet) weight}}{\text{radium-226 per gram of water}}$$

Algae were found to concentrate radium-226 within a range of 500 to 1,000. In the Animas River survey, background levels of radium-226 from unpolluted areas were found to be 4.5 picograms per gram of ash weight for algae, 3.5 picocuries for aquatic insects, and 0.44 picocuries for fish. The ash weight comprises 4 to 5 percent of the live weight. At a sampling station located approximately 15 miles downstream from milling operations, specimens taken revealed radium-226 concentrations as great as 152 picograms per gram ash weight for algae, 230 picograms for insects, and 24 picograms for fish. These values represent concentration factors of 844 for algae, 1,280 for insects, and 133 for fish; hence, graphically illustrate the accumulative abilities of algae and invertebrates.

Attached algae and selected benthic invertebrates have additional advantages over other stream organisms with regard to their selection as practical, long-term indicators of radioactivity pollution. Such advantages include ease of collection under field conditions and immobility.

CROPS

The effects on farm crops grown in radium-226 contaminated soils and/or irrigated with radium-226 polluted waters have been investigated.[46, 55, 61] In control studies, vegetables and fruits grown in soils having background radium-226 concentrations of 1.4 picograms per gram and irrigated with uncontaminated water, contained 2.0 picograms per kilogram wet weight. By contrast, produce grown on soils containing as high as 17 picograms per gram radium and irrigated with radium-polluted water, contained concentrations as great as 11 picograms per kilogram wet weight.

Of the plants studied, alfalfa samples concentrated radium-226 to a much greater degree when grown in the presence of radium-contaminated soil and water. In one area alfalfa had radium-226 concentrations ranging from 12.0 to 27.5 picograms per kilogram wet weight. In another area the concentration in two samples was 210 and 320 picograms radium-226 per kilogram wet weight. Milk samples from cows fed contaminated alfalfa were found to contain radium-226 concentrations as high as 5.0 picograms per liter.

Phosphate fertilizers contain trace amounts of natural radioactivity and were possible contributors to the radium-226 content of the crops. Investigations showed fertilizers to contain radium-226 concentrations as high as 21 picograms per gram. Alfalfa analysis showed that crops fertilized with radium-226 contaminated fertilizer contained somewhat higher amounts of radium-226 than those fertilized with uncontaminated fertilizer; however, a stronger effect was observed in crops irrigated with water containing radioactive elements.

With present day waste treatment practices it is unlikely that crop samples would be found to contain levels of radioactivity as high as previously mentioned. No crop samples have been analyzed in recent years, however, to substantiate a decrease in contamination as a result of waste treatment processes.

At the present time cattle are grazing near waste tailings areas in several locations where wind and water erosion has carried tailings material into pastures. The effect of ingesting contaminated pasture grass is unknown. Uptake of radioactivity by cattle may pass on to the general population through milk and meat consumption.

SECTION XII

STANDARDS FOR RADIOLOGICAL PROTECTION

Small amounts of a large variety of naturally-occurring radionuclides are always present in the environment, and are responsible for normal background radioactivity. Other radioactive materials are produced artificially through nuclear weapons testing and nuclear power reactors.

Water and comestibles normally contain an amount of radioactivity dependent upon their contact with radionuclides in the environment. On ingestion, radioactive substances are absorbed and become lodged in bones or other organs. The radionuclides emit alpha, beta, and gamma radiations that are injurious to body tissue above certain levels. The most hazardous of the radionuclides are naturally-occurring radium-226 and artificially-produced strontium-90. The maximum permissible standards for these elements are significantly lower than for other radionuclides.

The amount of strontium-90 in the environment is the result of past nuclear explosions and relatively constant; therefore, any reduction in the hazardous radioactive materials present must be from a reduction in radium-226 concentration. A significant amount of radium-226 released to the environment is a consequence of uranium milling and refining operations. The amount of radium-226 permitted in the environment is dependent on the amount of strontium-90 present. The ratio of radium-226 concentration to the maximum permissible concentration plus the ratio of strontium-90 to the maximum permissible concentration should not be greater than one.

Several guidance documents on radiation exposure for various radionuclides have been issued through the years. The trend in allowable exposure levels has been downward and will probably be further lowered in a document soon to be released. All of the documents contain maximum permissible concentrations or ranges for radium-226 and strontium-90.

INTERNATIONAL COMMISSION ON RADIOLOGICAL PROTECTION REPORT

For continuous occupational exposure, the ICRP report lists the maximum permissible concentration for dissolved radium-226 in water as 100 picocuries per liter and 300,000 picocuries per liter for suspended radium-226.[31] The concentrations are based on the assumption that an average person consumes 2.2 liters of water per day or its food equivalent.

The levels are reduced by a factor of 10 to give the maximum permissible concentration for an individual of 10 picocuries radium-226 per liter (22 picocuries per day). A factor of 30 is used to give the maximum permissible concentrations for the general public of 3.3 picocuries radium-226 per liter (7.3 picocuries per day). The limits apply only in the absence of other bone-seeking radionuclides such as strontium-90. In their presence the limits should be reduced accordingly. The report sets the limits of strontium-90 for the general public at 33 picocuries per liter--10 times higher than the limit for radium-226.

NATIONAL BUREAU OF STANDARDS HANDBOOK 69

An abridgement of the ICRP report, the handbook gives the recommendations of a group of experts, the National Committee on Radiation Protection and Measurements, on limits of radiation exposure.[30] The publication contains the same table of maximum permissible concentrations as the ICRP report and refers to the report for factors to reduce the occupational limits to levels considered safe for the general public. Body burdens and concentrations of radionuclides may be averaged over a period of one year.

FEDERAL RADIATION COUNCIL STANDARDS

The Federal Radiation Council was established to advise the President regarding radiation matters affecting health, to give guidance to all Federal agencies in the formulation of radiation standards, and to establish and execute programs of cooperation with states. The initial recommendations of the council were published in the Federal Register on May 18, 1960.[62] A second report offered specific guidance concerning exposure of population groups to four radionuclides, including radium-226 and strontium-90.[63]

The council deviated from the use of a single intake value for maximum permissible concentrations of radionuclides, and established three ranges of concentration with a graded scale of action for each range. It was reasoned that a single numerical concentration does not provide adequate guidance for taking appropriate action in all situations. The three ranges of intake are shown in Table 28. The ranges of transient rates of intake for the graded scale of action are shown in Table 29. The upper limit of Range II (Table 29) is based on an annual concentration considered as an acceptable risk for a lifetime.

Compared to the ICRP and NCRP recommended standard for radium-226 of 7.3 picocuries per day, an average of Range II (2 to 20 picocuries per day) is not greatly different. Use of the range concept prevents an alarmist interpretation of transient levels of exposure. The range allows a higher level of concentration without cause for concern, but also recommends surveillance and control at a level of two picocuries of radium-226, lower than the ICRP and NCRP recommendations.

USPHS DRINKING WATER STANDARDS--1962

The standards, guided by the Federal Radiation Council recommendations, state that water supplies shall be approved without further consideration of other sources of radioactivity when radium-226 and strontium-90 concentrations do not exceed three and 10 picocuries per liter, respectively.[64] Should the concentrations be exceeded, a water supply shall be approved after surveillance of total intakes of radioactivity from all sources indicates that such intakes are within the Range II limits of 2 to 20 picocuries per day of radium-226.

Table 28. GRADED SCALE OF ACTION

Ranges of transient rates of daily intake	Graded scale of action
Range I	Periodic confirmatory surveillance as necessary.
Range II	Quantitative surveillance and routine control.
Range III	Evaluation and application of additional control measures as necessary.

Table 29. RANGES OF TRANSIENT RATES OF INTAKE FOR USE IN GRADED SCALE OF ACTION
(picocuries per day)

Radionuclide	Range I	Range II	Range III
Radium-226	0- 2	2- 20	20- 200
Strontium-90	0-20	20-200	200-2,000

WATER QUALITY CRITERIA--FWPCA

The National Technical Advisory Committee on Water Quality Criteria, established February 27, 1967, assembled a comprehensive document on water quality requirements to be used as a basic reference by groups and agencies engaged in water quality studies and standard-setting activities.[65] The committee established standards for surface water criteria for public water supplies in two groupings. The first, permissible criteria, included those characteristics and concentrations of substances in raw surface water that allow for the production of a safe, clear, potable, aesthetically pleasing, and acceptable public water supply meeting the limits of the Public Health Service Drinking Water Standards of 1962 after treatment. The second, desirable criteria, included those characteristics and concentrations of substances in raw surface waters that represent high quality water in all respects for use as public water supplies. The standards for radioactivity and also for the most probable inorganic contaminants present in uranium mining and milling wastes are given in Table 30.

Table 30. SURFACE WATER CRITERIA FOR PUBLIC WATER SUPPLIES

Constituent	Permissible criteria	Desirable criteria
Radioactivity:	(picocuries per liter)	(picocuries per liter)
Gross beta	1,000	<100
Radium-226	3	<1
Strontium-90	10	<2
Inorganic chemicals:	(milligrams per liter)	(milligrams per liter)
Chloride	250.00	<25
Sulfate	250.00	<50
Nitrates	10.00	virtually absent
Arsenic	0.05	absent
Selenium	0.01	absent
Uranyl Ion	5.00	absent

During 1970, a number of questions were raised regarding the adequacy of radiation protection standards. To provide satisfactory answers to the queries, the National Academy of Sciences was asked to thoroughly review all pertinent, available scientific data and recommend changes deemed necessary. A report of findings by the organization has not been released. Additionally, an EPA working group reviewing drinking water standards is expected to publish revised standards with lowered radioisotope levels.

In summary, all documents concerning water quality standards for radioactive wastes are in relatively close agreement. The continuing trend in maximum permissible concentrations is downward. The limits of standards are considered to be maximums and every effort should be made by the uranium industry in pollution abatement programs to reduce the concentrations of pollutants released to the environment to as low a level as is possible, rather than discharging wastes to the preselected level of the standard. Abatement procedures have demonstrated that the level of concentration of radium-226 in river water can be maintained at less than one picocurie per liter.

SECTION XIII

TESTING AND MONITORING PROGRAMS

Although considerable testing and monitoring for pollution caused by uranium mining and milling activities has been conducted in the Colorado River Basin, other uranium-producing areas have received scant attention. The information gained in the Colorado River Basin studies has been invaluable in identifying problem areas within the industry.

To adequately define pollution due to radioactivity, analysis for radium-226 is necessary since this radionuclide is considered to be the most hazardous of the naturally-occurring radionuclides and has been given the lowest maximum permissible concentration. As a general rule, when radium-226 concentrations are found to be below the standard limit, all other radionuclide concentrations will be below their standard limit. Uranium analysis should also be performed. In addition to the specific analyses mentioned, gross alpha and gross beta activity should be measured to insure that other radionuclide concentrations are low. In instances of higher gross alpha or gross beta activities than would be expected from the radium-226 and uranium content, analyses for other specific radionuclides are indicated: thorium isotopes, lead-210, polonium-210, and strontium-90.

While radiological pollution has been the major concern from the uranium industry wastes, analyses for other inorganic elements should not be neglected. The frequency of analysis for chlorides, sulfates, nitrates, arsenic, selenium, and molybdenum should depend on the levels found in previously analyzed samples.

The major problems encountered in testing and monitoring programs are the lack of sufficient data, the intermittance of data, and the reliability of data obtained. Background data are not available for many areas to utilize in assessing the degree of pollution. Split samples analyzed by different laboratories have shown discrepancies, indicating the need for improved quality control measures to assure the validity of the results.

All companies involved in milling and refining of radioactive materials must meet certain requirements. The companies are licensed by the Atomic Energy Commission or by states designated as agreement states. Agreement states have assumed all licensing, record keeping, and inspection responsibilities from the Atomic Energy Commission. Of the seven states in which milling activities are presently being conducted, Colorado, Texas, and Washington are agreement states.

The licensing requirements are set forth in the Code of Federal Regulations, Title 10, Part 20 (10 CFR 20).[66] One requirement of the regulations is that the company conduct a continuous sample monitoring program to verify that the mill is operating within defined limits set forth in the license. The records must be kept available for periodic inspection by the Atomic Energy Commission or the agreement states.

The 10 CFR 20 limit for soluble radium-226 that may be discharged to an unrestricted area is 30 picocuries per liter and for insoluble or suspended radium-226, 30,000 picocuries per liter. The section further states, "In addition to limiting concentrations in effluent streams, the Commission may limit quantities of radioactive materials released in air or water during a specified period of time if it appears that the daily intake of radioactive material from air, water, or food by a suitable sample of an exposed population group, averaged over a period not exceeding one year, would otherwise exceed the daily intake resulting from continuous exposure to air or water containing one-third the concentration of radioactive materials specified . . . ." For soluble radium-226, the amount would be one-third of 30, or 10 picocuries per liter.

The State of Colorado operates a radiochemistry laboratory and conducts a monitoring program. The State has cooperated through meetings and sampling surveys with Federal agencies to assess pollution effects on stream waters. Other states perform alpha and beta analyses on samples and occasionally gamma scans. Most states are not equipped to perform radioactive analyses of the radionuclides in the uranium mill waste discharge. As laboratory apparatus and equipment needed for the analyses are quite expensive for the number of analyses required in a monitoring program, the states often enlist the aid of a Federal agency.

Federal agencies have been involved in testing and monitoring activities concerned with the uranium industry since the 1950's. Both short-term (one year or less) and continuous long-term monitoring activities have been carried out in the Colorado River Basin.[29, 46, 55] A Radium Monitoring Network was established for the basin and samples for analysis collected periodically throughout the basin by the Colorado River Basin Laboratory in Salt Lake City, Utah. The first monitoring station was established in January, 1961, with the number eventually increasing to a maximum of 27 stations. The network was discontinued in September, 1969, with 19 sampling stations. Data from the sampling stations were assembled in 16 Radium Monitoring Network Data Releases. In October, 1969, many network stations were transferred into a Water Quality Surveillance System along with new stations. The frequency of collection at the 25 stations varied from one week to semi-annually. Data from the stations are stored by computer in the Storet system.

Sample analyses for the Colorado River Basin Project consisted of gross alpha, gross beta, gross gamma scans, radium-226, natural uranium, thorium, lead-210, polonium-210, and strontium-90. Not all parameters were measured on every sample; however, gross alpha, gross beta, radium-226, and natural uranium were routinely performed. Some of the analytical methods were developed or perfected in the Colorado River Basin Laboratory;[67, 68, 69] others were taken from Standard Methods.[70] In addition to long-term sampling programs, special surveys were conducted. To obtain a comprehensive overview of the effects of uranium milling waste products on the environment, many types of

samples other than water were analyzed, including river and reservoir sediments, topsoil, water treatment plant filter sands, aquatic biota, crops, and milk.

SECTION XIV

TECHNOLOGICAL ADVANCES IN THE URANIUM INDUSTRY

No major advancements have been made in uranium mining, milling, and waste treatment during the past thirteen years; however, research has produced new and improved methods that may eventually be used in the uranium industry. The methods will be discussed in detail below.

PHYSICAL UPGRADING OF LOW GRADE ORES

As the demand for uranium increases, processing of lower grade ores will be required. Processing of larger quantities of ore will result in excessive consumption of leaching reagents to produce the same amount of uranium concentrate. The leaching and neutralization process is the most costly phase of the milling processes as seen in Table 31.[71] Reduction of the volume of ore required to produce the same amount of uranium would result in a tremendous cost savings and prevent a larger waste disposal problem.

Table 31. DIRECT OPERATING COSTS OF URANIUM EXTRACTION IN THE U.S.A.

Process[a]	Percent of cost[a]
Crushing	6
Grinding and thickening	5
Leaching and neutralization	43
Liquid/solid separation	6
Extraction and purification	8
Precipitation, drying, and packaging	9
Services (effluent treatment, administration, etc.)	23

[a] Intermediate-sized mill (1,500-2,000 ton/day) operating on 5 lb/ton ore, with solvent extraction.

Methods of lump-ore sorting by machines may prove useful in reducing the bulk of ore processed fivefold with a uranium recovery rate of 90 to 95 percent. Radiometric, conductimetric, and optical detection systems have been used for ore sorting. Another promising method is high-intensity wet magnetic separation utilizing superconducting magnets.

More automation of equipment in the ore separation and grinding processes would prevent exposure of employees to areas of high dust concentration. Automation in the uranium concentrate packaging process would also eliminate another source of high dust exposure.

IMPROVED URANIUM EXTRACTION PROCESSES

An improved Eluex process has been developed that differs from existing ones in that a stage of uranium solvent extraction is coupled with each stage of resin elution rather than the elution and solvent extraction operations being conducted separately.[72] The improved system will reduce the number of stages, retention time, and resin inventory to about one-fourth or one-fifth that in existing circuits. Such improvements in circuit design may influence the design of milling processes in mills constructed in the future. From a waste treatment standpoint, processes involving maximum recycle of tailings solution should be one of the major goals.

Extraction of uranium from mine water by ion exchange techniques has been in use in some uranium mining areas since 1963. Mine water contains sufficient uranium to warrant recovery from an economical standpoint. An improved countercurrent ion exchange process has been developed for recovery of uranium from natural mine waters.[27] The improved technique reduced resin inventory requirements by 70 percent. Additional savings in equipment and labor costs would be realized by utilization of this process.

EXTRACTION OF URANIUM FROM SEAWATER

Uranium is present in seawater in concentrations of 2.9 to 3.3 micrograms per liter. Seawater is the lowest grade but the most abundant source of uranium. Of the trace metals in seawater, uranium is considered to be the only one present in sufficient quantities to warrant economic extraction in the foreseeable future.[2, 73] Studies have shown that 82 percent of the uranium may be rapidly extracted by a flotation technique. While it is unlikely that this source of uranium would be considered unless ore reserves become depleted, the technology for extraction has been developed.

Other studies of uranium recovery from seawater have been made that consist of concentrating uranium on adsorber beds in a coastal lagoon where a constant tidal flow can be maintained across the beds.[2] Hydrous titanium oxide was found to be a suitable adsorber. Ammonium carbonate proved to be the best solvent for extraction of uranium from the beds, resulting in an 80 percent removal from one pass of solution. The life of the beds is estimated at less than ten years. Final estimated cost of processed material was in the range of $27 to $43 per pound of uranium oxide. The cost is high for present uses of uranium, but in the event of a shortage, or breeder reactors are placed into commercial use, the cost might prove reasonable.

UNDERGROUND URANIUM EXTRACTION USING NUCLEAR EXPLOSIVES

Nuclear explosives have been used to increase the recovery of underground resources. Underground nuclear explosions conducted under the Plowshare program have increased the rate of gas production in underground formations by increasing the porosity and permeability of the formation. If nuclear explosive projects prove to be safe and economically competitive with conventional fracturing methods, the method could possibly prove useful in combination with underground solution mining of uranium ore bodies.[74] The problem of radioactive contaminated waste would be increased, however, due to the production of artificial radionuclides.

RADIUM REMOVAL FROM URANIUM MILL EFFLUENTS AND TAILINGS SOLIDS

Ion exchange techniques for radium removal from acid waste liquors have been studied.[42] Of thirteen solids tested, ten adsorbed more than 75 percent of the radium from a simulated lime-neutralized feed. The most effective solid, a commercially synthetic zeolite, removed 96 percent of the radium from solution. A brief study was made of the elution of radium from the ion exchangers. Ammonium nitrate and ammonium chloride proved to be the most efficient reagents with elution efficiencies as high as 90 percent. Greater than 97 percent of the radium was removed from the pregnant solution by barium sulfate precipitation. The eluted resin was reused for another adsorption cycle of radium with the same efficiency.

A study conducted by the Bureau of Mines indicated that 90 percent of the radium could be removed from tailings solids by a versenate leaching process, but the cost was estimated to double the price of uranium.[33] Radium is significantly leached from tailings solids by water or barium chloride solution.[53] Removal of radium from waste tailings solids would make disposal much less of a problem.

SECTION XV

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